

Atmospheric Mercury Measurements and Modeling at NOAA's Air Resources Laboratory

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<http://www.arl.noaa.gov/mercury.php>

Presentation at:
NCAS Annual Science Meeting
April 15, 2010
at the NOAA Silver Spring Campus



1. Measurements

- A. Site Locations and Settings
- B. Current Suite of Measurements
- C. Intensive (this Summer)
- D. Data – some examples

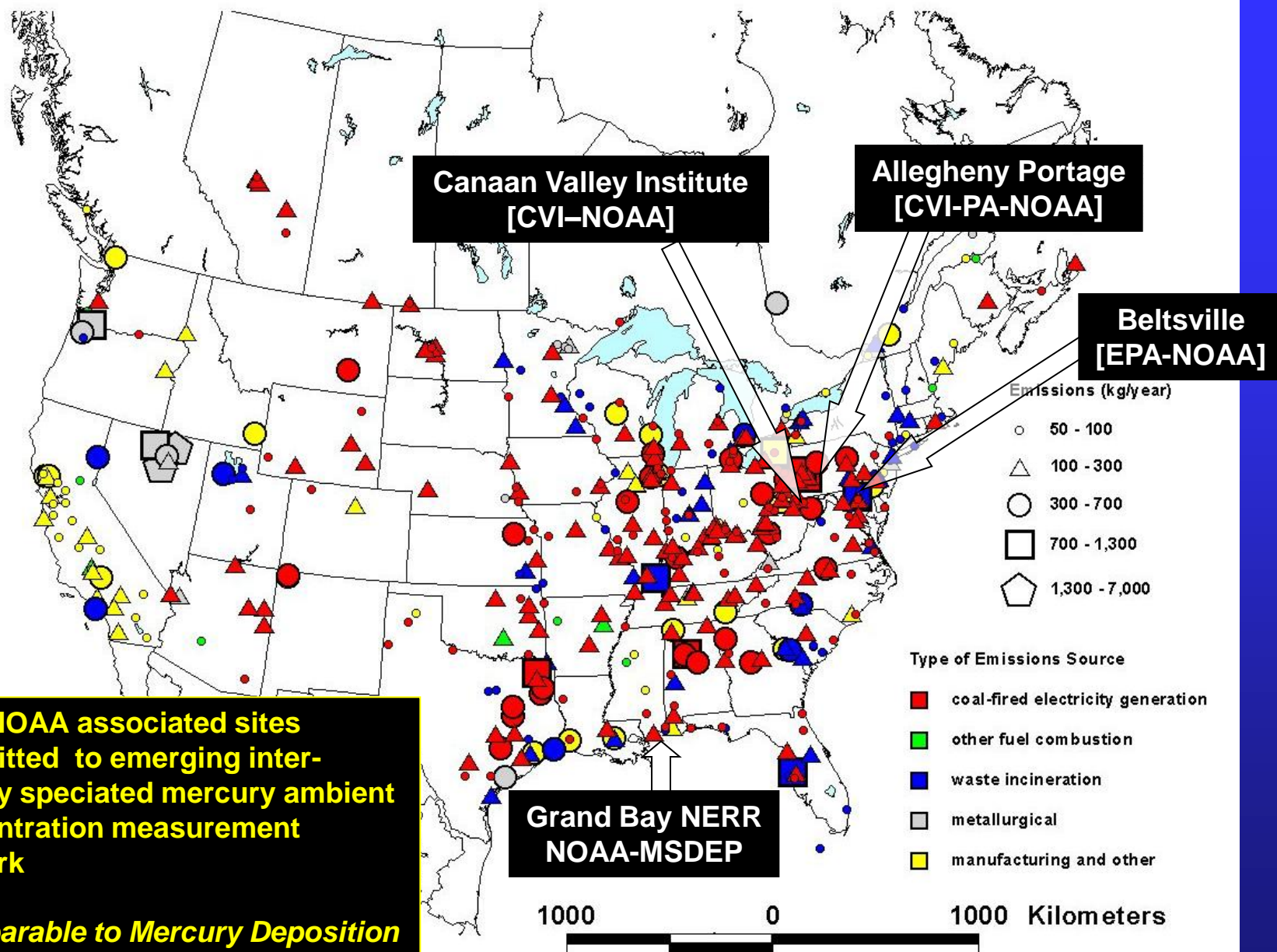


2. Modeling

- A. Episodes for Model Evaluation
- B. Emissions and Met Data -- Fine-Scale
- C. Evolution of the HYSPLIT-Hg Model
- D. Upcoming Great Lakes Project

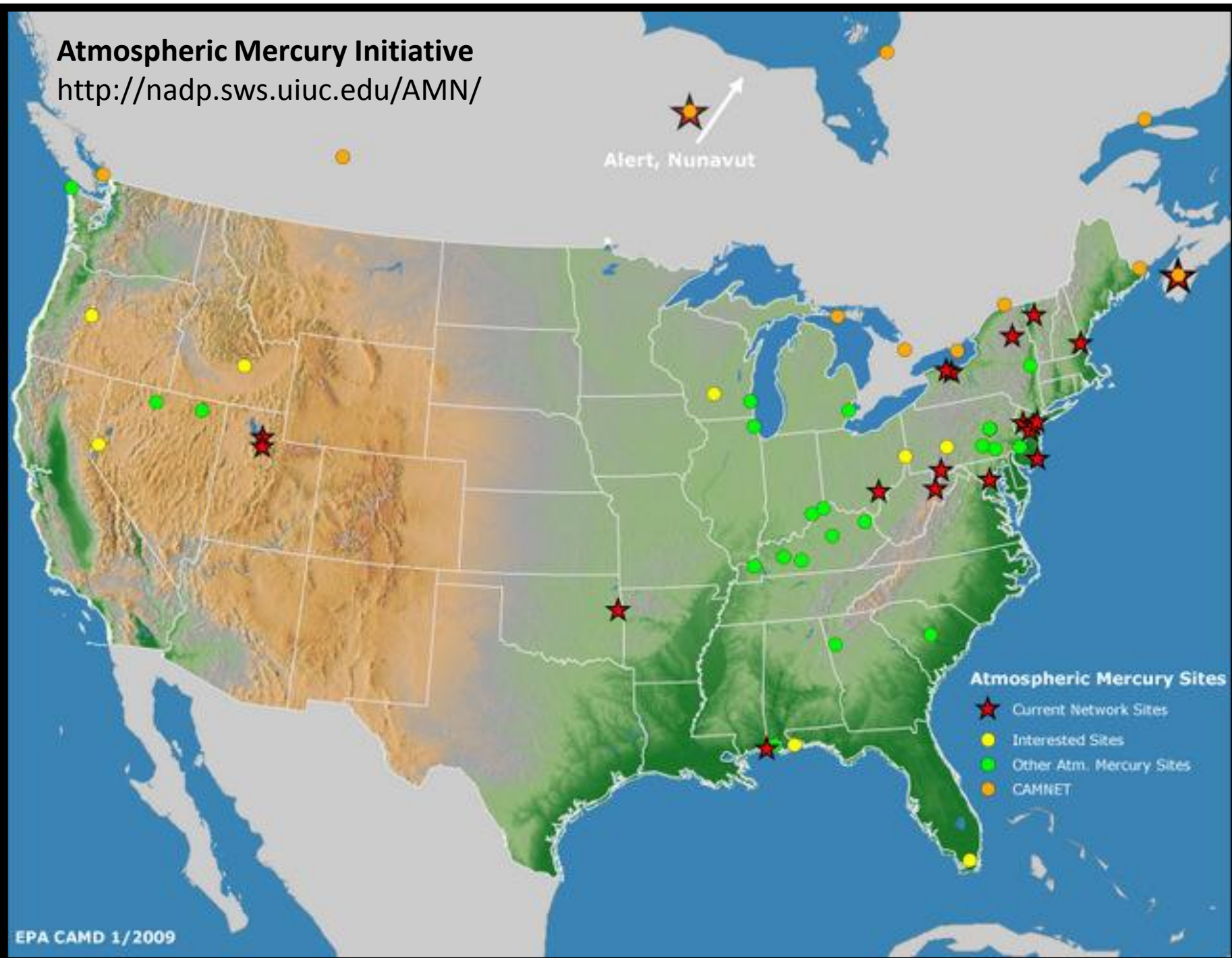
Four NOAA associated sites committed to emerging inter-agency speciated mercury ambient concentration measurement network

(comparable to Mercury Deposition Network (MDN) for wet deposition, but for air concentrations)



Atmospheric Mercury Initiative

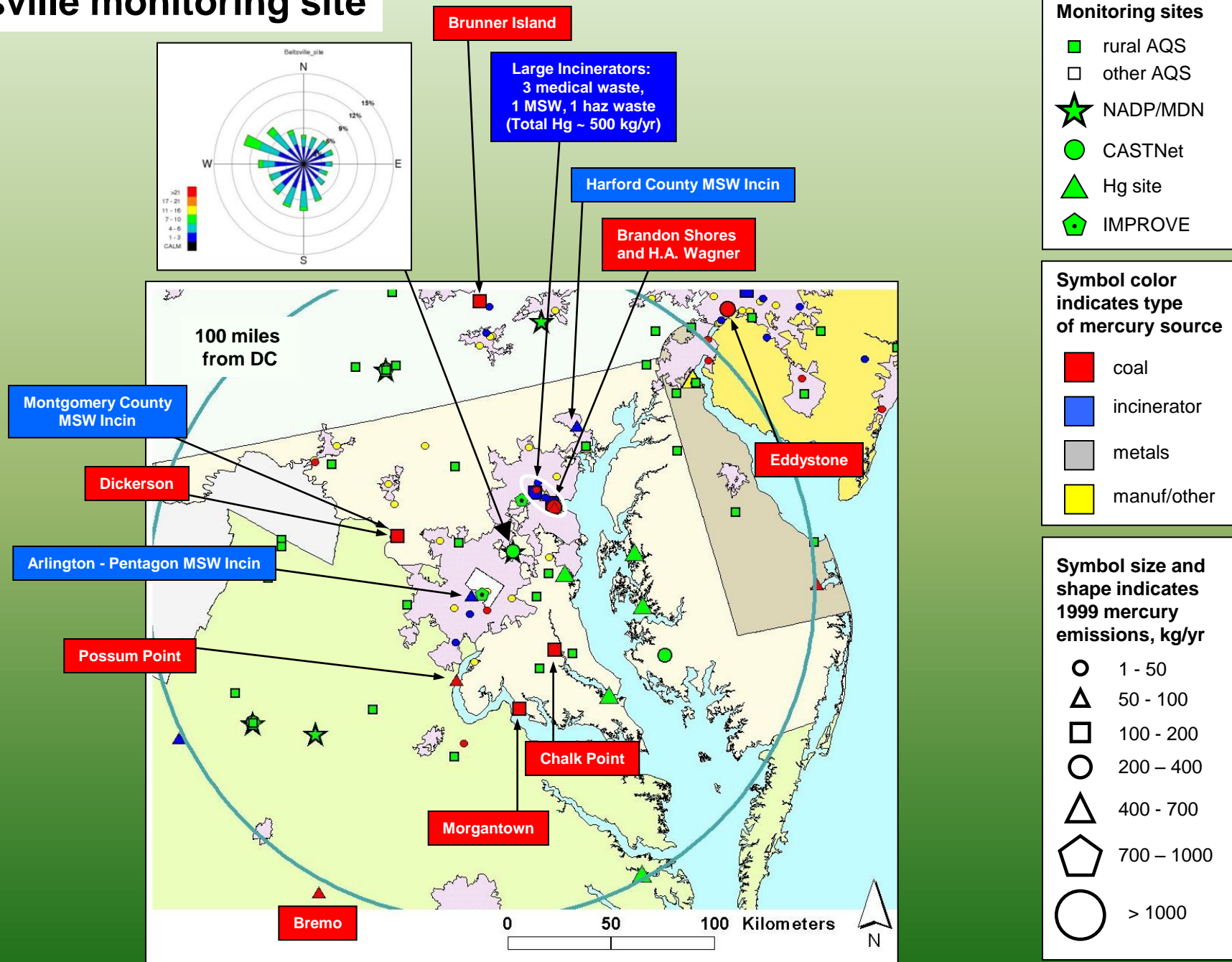
<http://nadp.sws.uiuc.edu/AMN/>

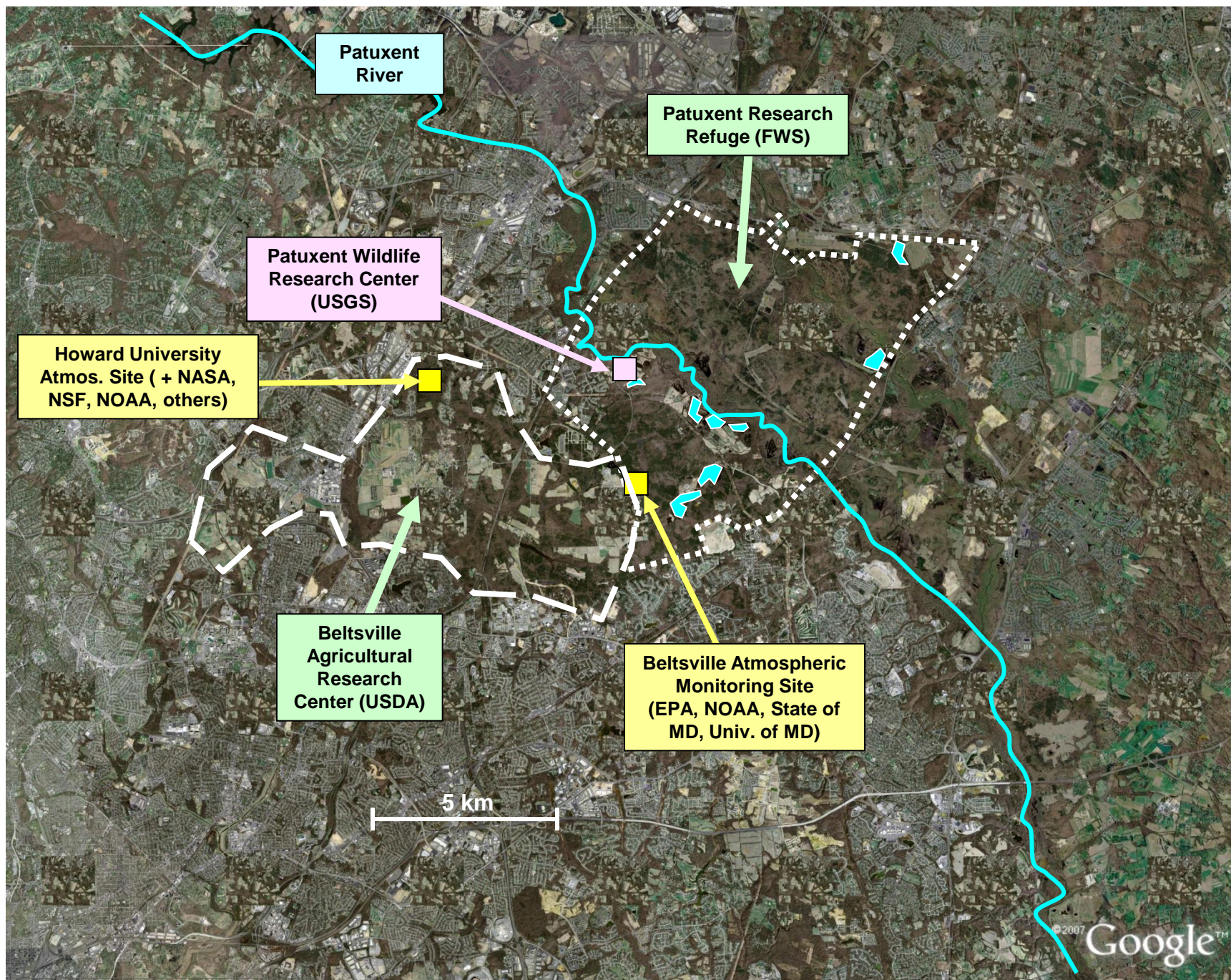


Beltsville, Maryland

**Co-located with the EPA's CASTNet site,
and the Beltsville NTN and MDN sites**

Beltsville monitoring site





Atmospheric Mercury Measurement Site at Beltsville, MD



ARL's Winston Luke working with RGM and Hg(p) collectors



Precipitation measurements (left to right): Mercury Deposition Network, Major Ions (e.g. "acid rain"), Precipitation Amount



mercury and trace gas monitoring tower (10 meters)



Top of tower (close-up) with two sets of RGM and Hg(p) collectors

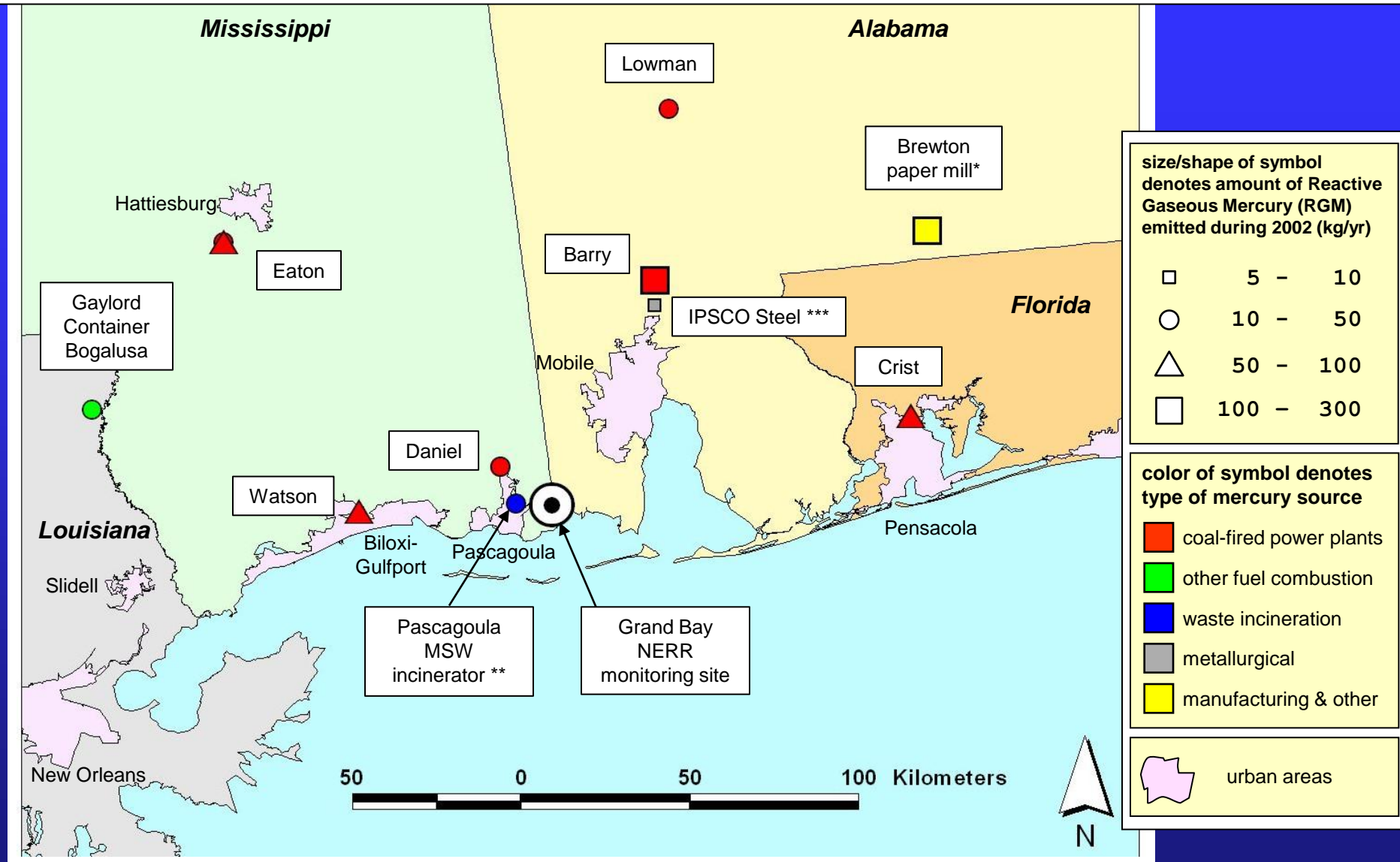


After RGM and Hg(p) is collected, it is desorbed and analyzed inside the trailer, along with Hg(0)

Grand Bay National Estuarine Research Reserve (NERR), Mississippi

**Co-located with the MS-DEQ / EPA's
precipitation measurements sites
(NTN, MDN, and trace metals)**

Grand Bay NERR sampling site, with large point sources of Reactive Gaseous Mercury in the region, based on the EPA's 2002 National Emissions Inventory



* Brewton paper mill mercury emissions included in 2002 NEI, but do not appear to be in 2000-2008 TRI

** Pascagoula MSW incinerator mercury emissions included in 2002 NEI but incineration ceased in Jan 2001

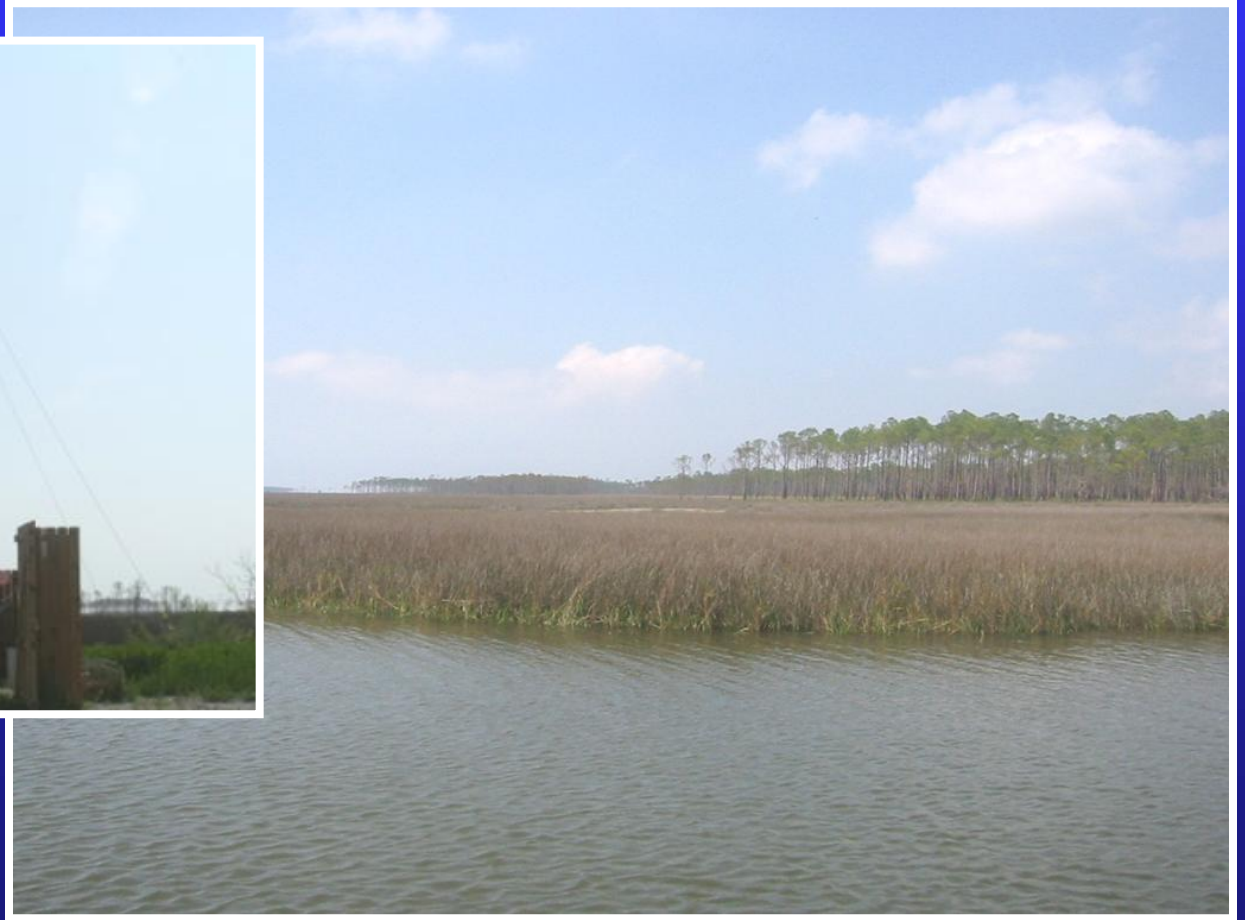
*** Ipsco Steel had significant mercury emissions in 2002 NEI, but negligible emissions reported in 2008 TRI

Current Location of Site

view from top of the tower



mercury and trace gas
monitoring tower
(10 meters)



“Hmmm...maybe it would work better if we have the tower go vertical?”



Winston Luke (Principal Investigator, NOAA – Air Resources Laboratory) and **Jake Walker** (site operator, Grand Bay National Estuarine Research Reserve)

Current Atmospheric Measurements at the Grand Bay NERR

Elemental mercury (two instruments)	}	“Speciated” Atmospheric Mercury Concentrations
Fine particulate mercury (two instruments)		
Reactive gaseous mercury (two instruments)		
Sulfur dioxide	}	Trace gases and other measurements to help understand and interpret mercury data
Ozone		
Carbon Monoxide		
Nitrogen Oxides (NO, NOy)		
Aerosol Black Carbon		
Wind speed, Wind Direction	}	Meteorological Data
Temperature, Relative Humidity		
Precipitation Amount		



Instrumentation inside the trailer at the Grand Bay NERR site



Timeline of Site Activities at the Grand Bay NERR



Sept 2006:
Speciated Hg,
SO₂, O₃, and CO
measurements begin
at “inland” site

2005-2006:
site selection

Aug 21 – Oct 5, 2008:
site shut down due to
threat of hurricanes

Summer 2010:
Field Intensive
(being planned)

Jan 2008:
NO/NO_y
added

Jan 2010:
Black
Carbon
added

Feb 2007: Meteorological
measurements added

Oct 2007:
Move to “coastal”
site near Pavillion;
2nd Tekran speciated Hg
measurement suite added

2010: Wet
Deposition
measurements
being added



Wet Deposition Measurements Being Added in 2010
by the Mississippi Department of Environmental Protection
(Henry Folmar, Becky Comyns, others), with funding from the EPA

Precipitation	Continuous digital measurement of precipitation amount
Major Ions pH, SO_4^{-2} , NO_3^- , PO_4^{-3} , Cl^- , NH_4^+ , Ca^{+2} , Mg^{+2} , K^+ , Na^+	Weekly measurements of concentrations in precipitation (NADP-NTN)
Total Mercury	Weekly measurements of concentration in precipitation (NADP-MDN)
Methyl Mercury	Monthly measurements of concentration in precipitation (composite)
Selected Trace Metals As, Cd, Cr, Cu, Pb, Ni, Se, Zn	Weekly measurements of concentrations in precipitation

Canaan Valley, West Virginia

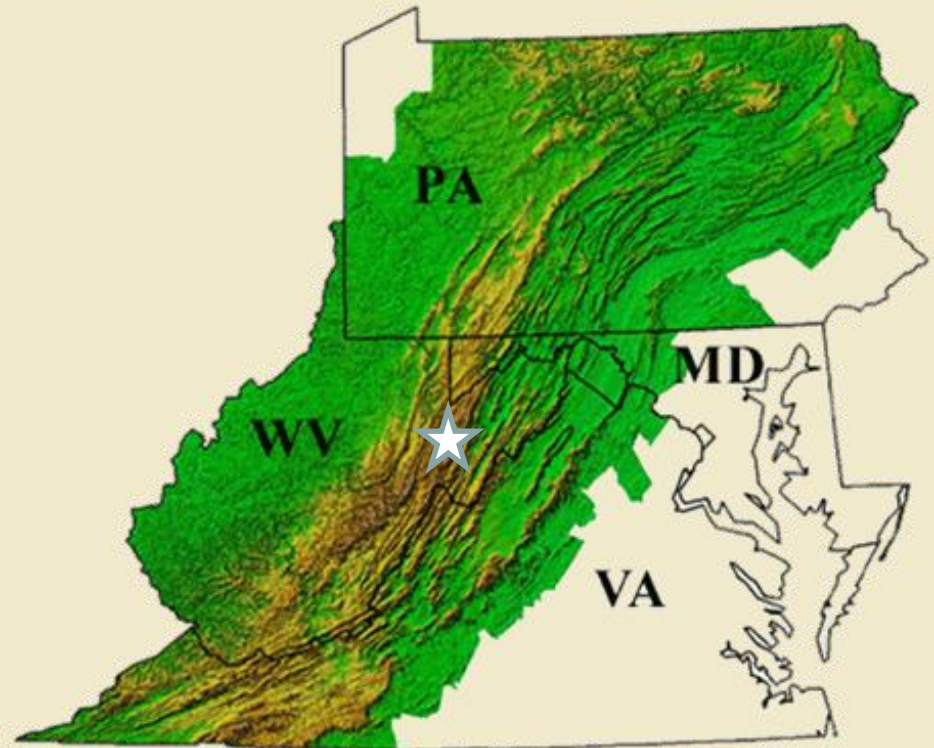
**Operated by Steve Brooks
of the Canaan Valley Institute,
who also runs an AIRMon-Wet site,
a new MDN site, and carries out
several other Hg measurements as well**



Canaan Valley Institute

CVI's Research Area is
the Mid-Atlantic
Highlands

NOAA performs the
Mercury and Air
Quality Studies



Canaan Valley non-Network Mercury Measurements

- **Continuous gaseous elemental mercury surface fluxes (modified-Bowen ratio)**
- **Weekly total mercury snow pack and stream outflow (calibrated stream gauge)**
- **Monthly groundwater**
- **Total mercury in Throughfall (event/campaign)**
- **Weekly Cat-ion exchange membrane surrogate surfaces**

Canaan Valley Hg Site

Ultrasonic anemometer
for wind turbulence

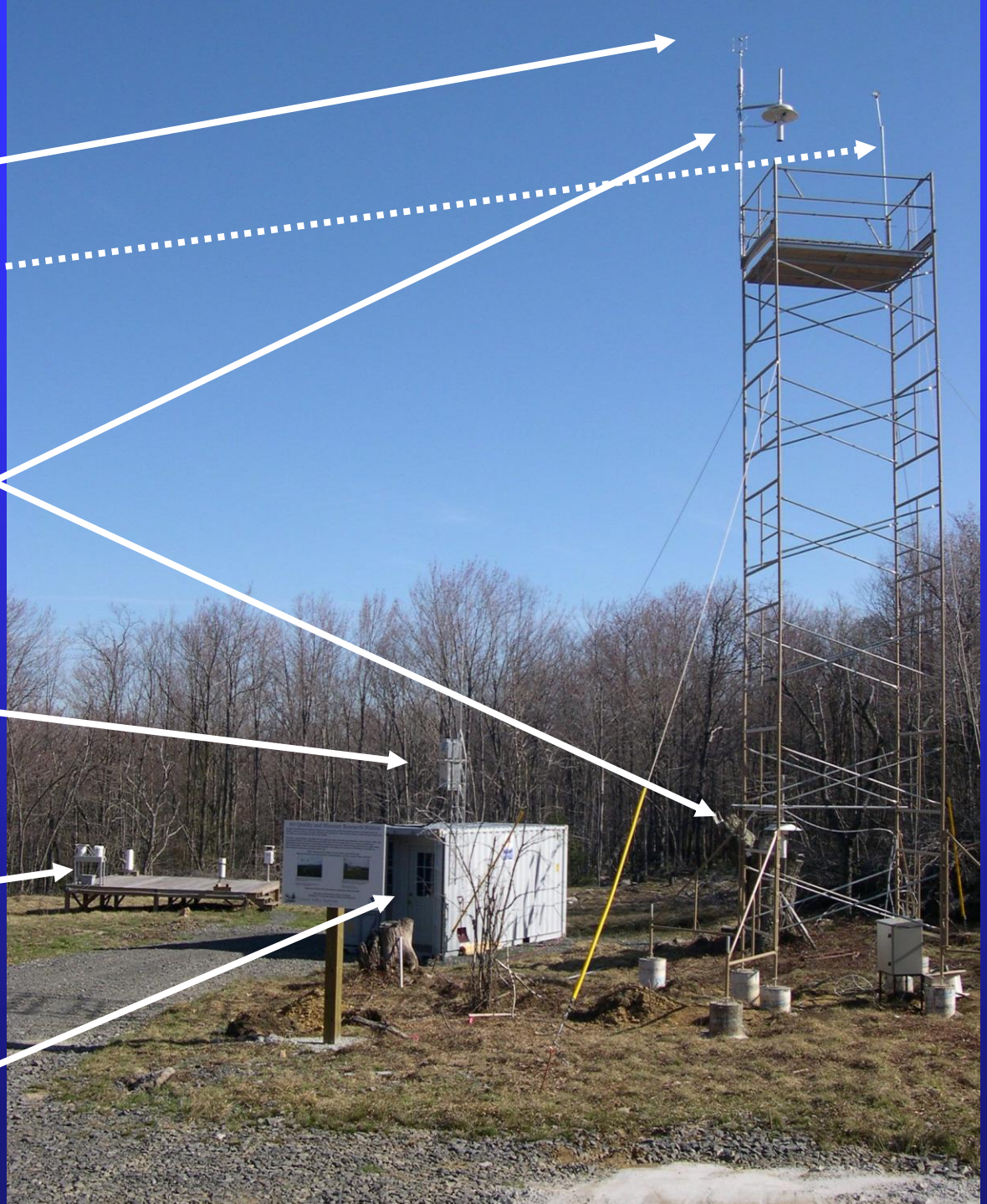
Charge dissipater for
Lightning protection

Vertical profilers for mercury
Air-surface exchange

Mercury chemical
composition sensors

Rain and Snow collectors for
Mercury and Acid Rain

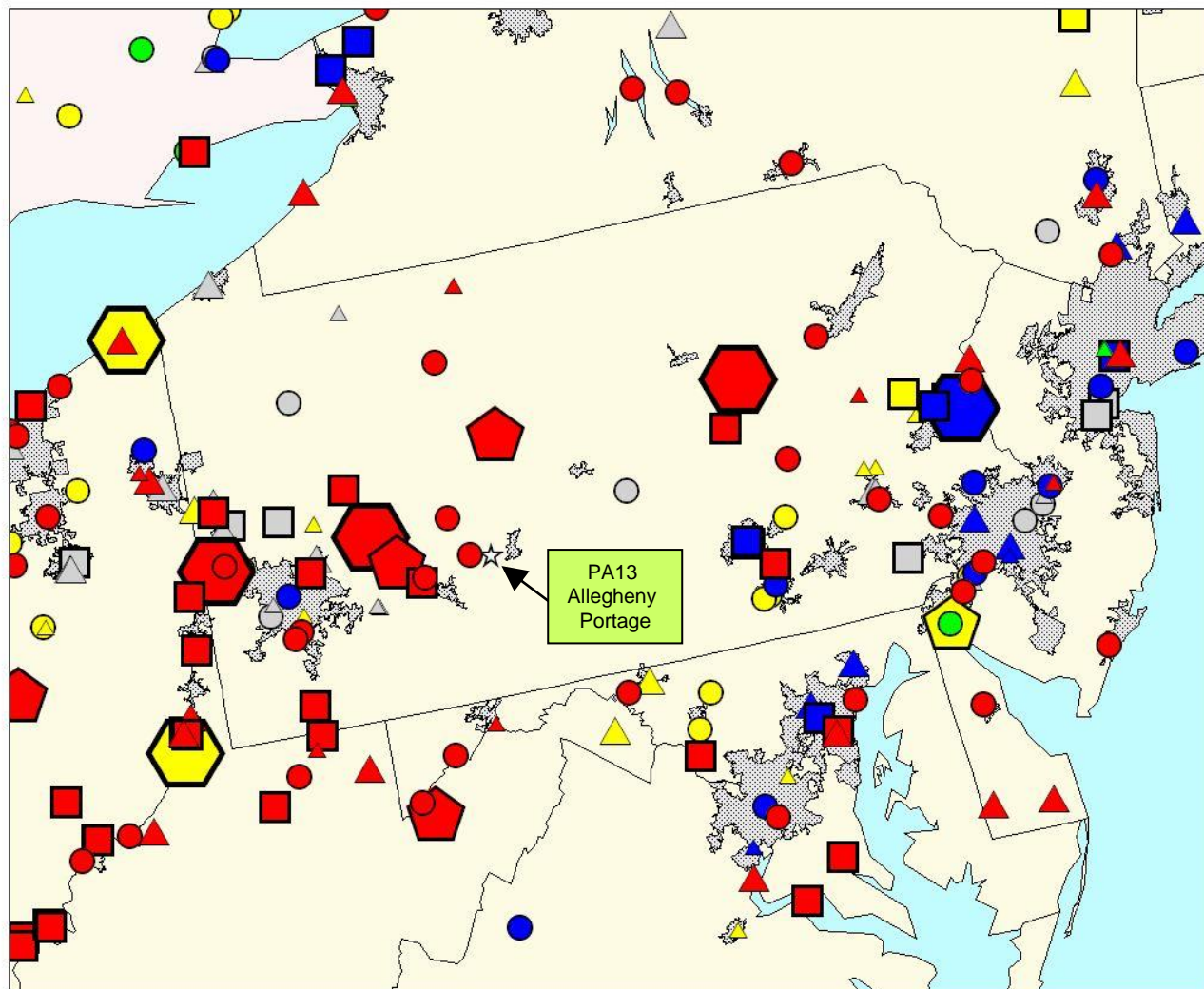
Shelter for sensors measuring
mercury, ozone, carbon
dioxide and other trace gases



Allegheny Portage Railroad National Historic Site, Pennsylvania

**Co-located with the MS-DEQ / EPA's
precipitation measurements sites
(NTN, MDN, and trace metals)**

Total mercury emissions from large point sources based on USEPA 2002 National Emissions Inventory (NEI)



Color of symbol indicates
type of emissions source

- Coal-Fired Power Plant
- Other Fuel Combustion
- Waste Incineration
- Metallurgical
- Manufacturing

Size/shape of symbol
indicates amount of
mercury emitted

- △ 5-10
- 10-50
- △ 50-100
- 100-300
- ⬡ 300-500
- ⬢ 500-1000
- △ 1000-3000

☆ Pa13 (Allegheny Portage)

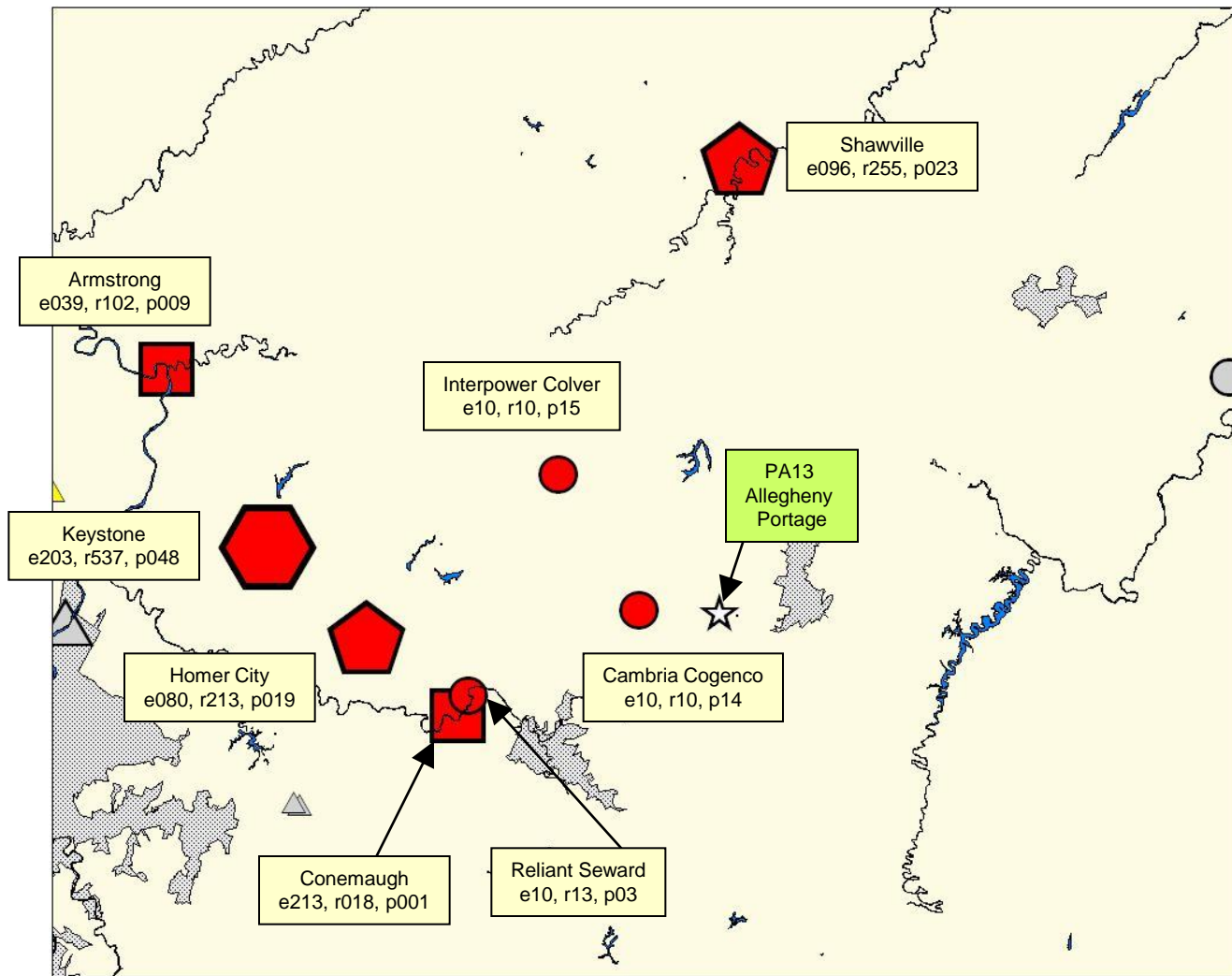
Urbanized areas

100 0 100 200 Kilometers



Total mercury emissions from large point sources based on USEPA 2002 National Emissions Inventory (NEI)

Numbers in facility labels are kg/yr emissions of elemental, reactive gaseous, and particulate mercury, respectively



Color of symbol indicates type of emissions source

- Coal-Fired Power Plant
- Other Fuel Combustion
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Size/shape of symbol indicates amount of mercury emitted

- △ 5-10
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- △ 50-100
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- △ 1000-3000

☆ Pa13 (Allegheny Portage)

Urbanized areas

25 0 25 50 Kilometers





PA13
Allegheny
Portage

Gallitzin

Hoguetown

Luckett

Sankertown

Cresson

Dysertown

Lilly

MDN PA 13

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Image USDA Farm Service Agency
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Google

1465 m
Imagery Dates: Jun 7, 2005 - Aug 5, 2006

lat 40.433433° lon -78.563466° elev 759 m

Eye alt 4.91 km



National Atmospheric Deposition Program

NADP/MDN Monitoring Location PA13

Station	Allegheny Portage Railroad National Historic Site (PA13)
Location	Cambria County, Pennsylvania
Dates of Operation	1/7/1997 - Present
Latitude	40.457
Longitude	-78.56
Elevation	739 meters
USGS 1:24000 Map Name	Cresson, 1981
Operating Agency	Allegheny Portage Railroad National Historic Site
Sponsoring Agency	Pennsylvania Department of Environmental Protection



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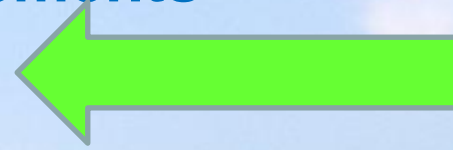
Summary of NOAA ARL Mercury Measurement Sites

● NOAA-led measurement
● Co-located measurement

Site	Collaborators / Co-locators	Ambient Air Measurements						Precipitation			Dry Deposition		Other
		Mercury Speciation	SO ₂	O ₃	NO/NO _y	CO	Carbon black	Major Ions (NTN)	Mercury (MDN)	Trace Metals	Surrogate Surface	Throughfall	Meteorology
Beltsville (MD)	<ul style="list-style-type: none"> PI = Winston Luke (NOAA) EPA Clean Air Markets Division Univ of Maryland Maryland DNR MACTEC USGS 	●●	●	●	●	●		●	●			●	●
Grand Bay (MS)	<ul style="list-style-type: none"> PI = Winston Luke (NOAA) Grand Bay NERR MS Dept Envr Quality U.S. EPA U.S. Fish & Wildlife Agency 	●●	●	●	●	●	●	●	●	●			●
Canaan Valley (WV)	<ul style="list-style-type: none"> PI = Steve Brooks (CVI/NOAA) Canaan Valley Institute Univ Md Frostburg Appalachian Lab USGS 	●		●				●	●		●	●	●
Allegheny Portage (PA)	<ul style="list-style-type: none"> PI = Steve Brooks (CVI/NOAA) Canaan Valley Institute Pennsylvania DEP National Park Service 	●						●	●				

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Plans for Atmospheric Field Intensive, July-Aug 2010 at the Grand Bay NERR

Ground-Based Measurements

(ongoing) speciated mercury, trace gas, black carbon, and meteorological measurements

- Winston Luke and Paul Kelley (NOAA ARL)
- Jake Walker (Grand Bay NERR)

(ongoing) wet deposition: major ions, mercury, methylmercury, metals
Will try to switch to event-based during intensive

- Mississippi State Dept of Env Protection/EPA
- Jake Walker (Grand Bay NERR)

ambient concentrations of BrO at the surface via Chemical Ionization Mass Spectrometry (possibly other Br compounds, e.g., Br₂, BrCl, and HOBr)

- Greg Huey (Georgia Tech)

isotopic mercury analysis of event-based precipitation and aerosols

- Bill Landing, Flip Froelich (Florida State Univ)

trace metal analysis of size-segregated aerosol
Spring 2010 and possibly during intensive

- Mark Engle (USGS)

Aircraft and Above Surface Measurements

aircraft flights measuring concentrations of Hg⁰ (Tekran), total and “speciated” RGM (coated/uncoated denuders), O₃, SO₂, and particle count

- Stephen Corda, John Muratore, & colleagues (Univ. of Tennessee Space Institute – UTSI)
- Hynes and Swartzendruber (Univ of Miami)
- Luke and Kelley (NOAA ARL)

vertical distribution of O₃ and met data above the site (ozonesondes)

- Luke and Kelley (NOAA ARL)
- Jake Walker (Grand Bay NERR)

Details are still being worked out...

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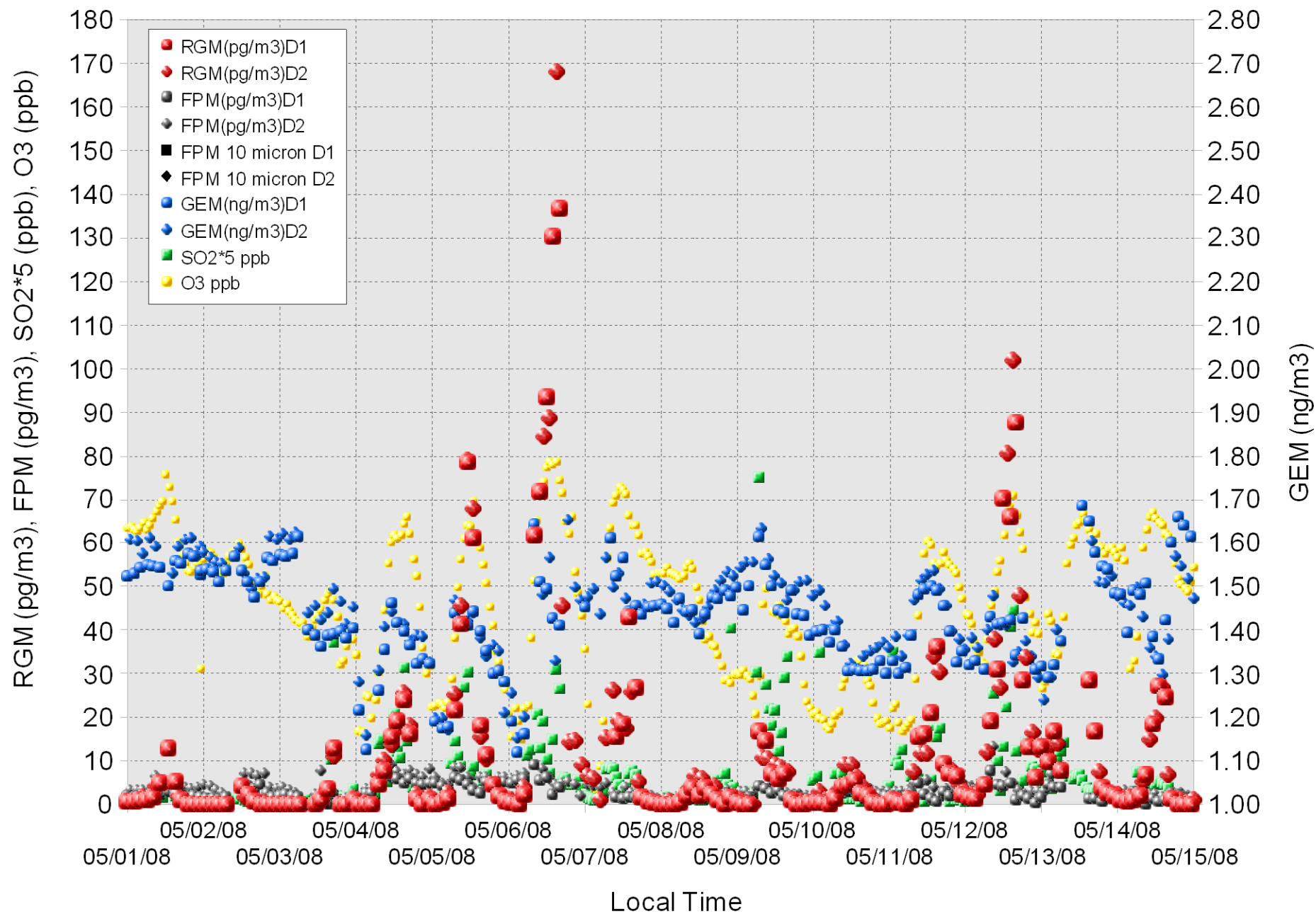


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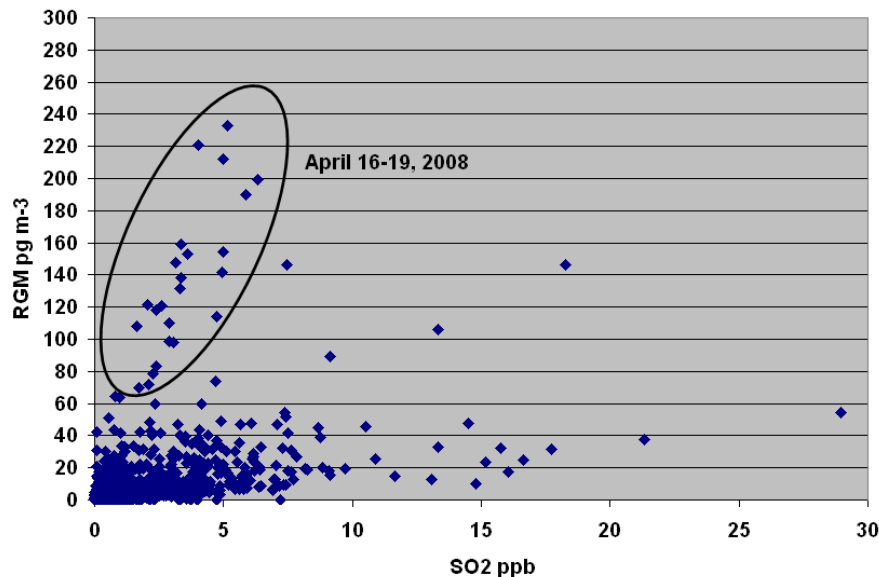
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Courtesy of Winston Luke and Paul Kelley (NOAA ARL) and Jake Walker (Grand Bay NERR) (Preliminary Values)

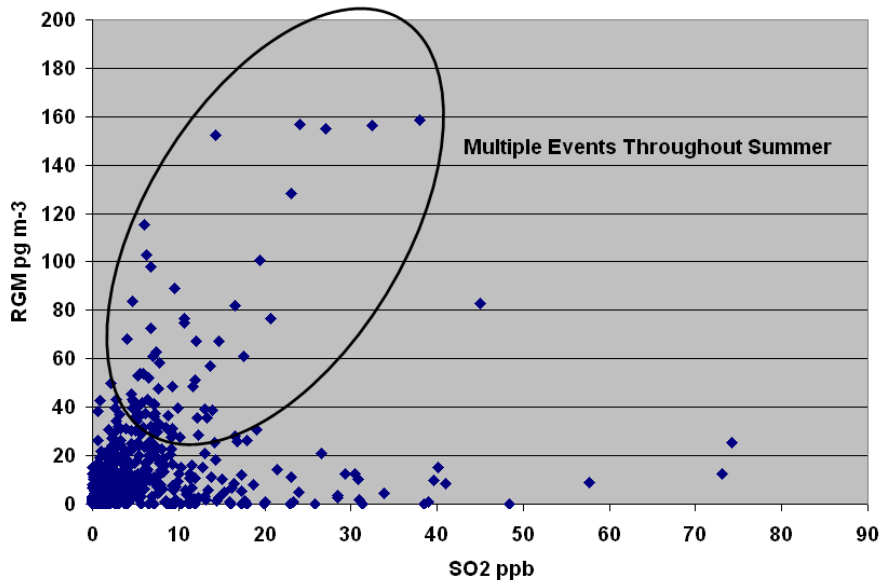


RGM vs SO₂ Spring



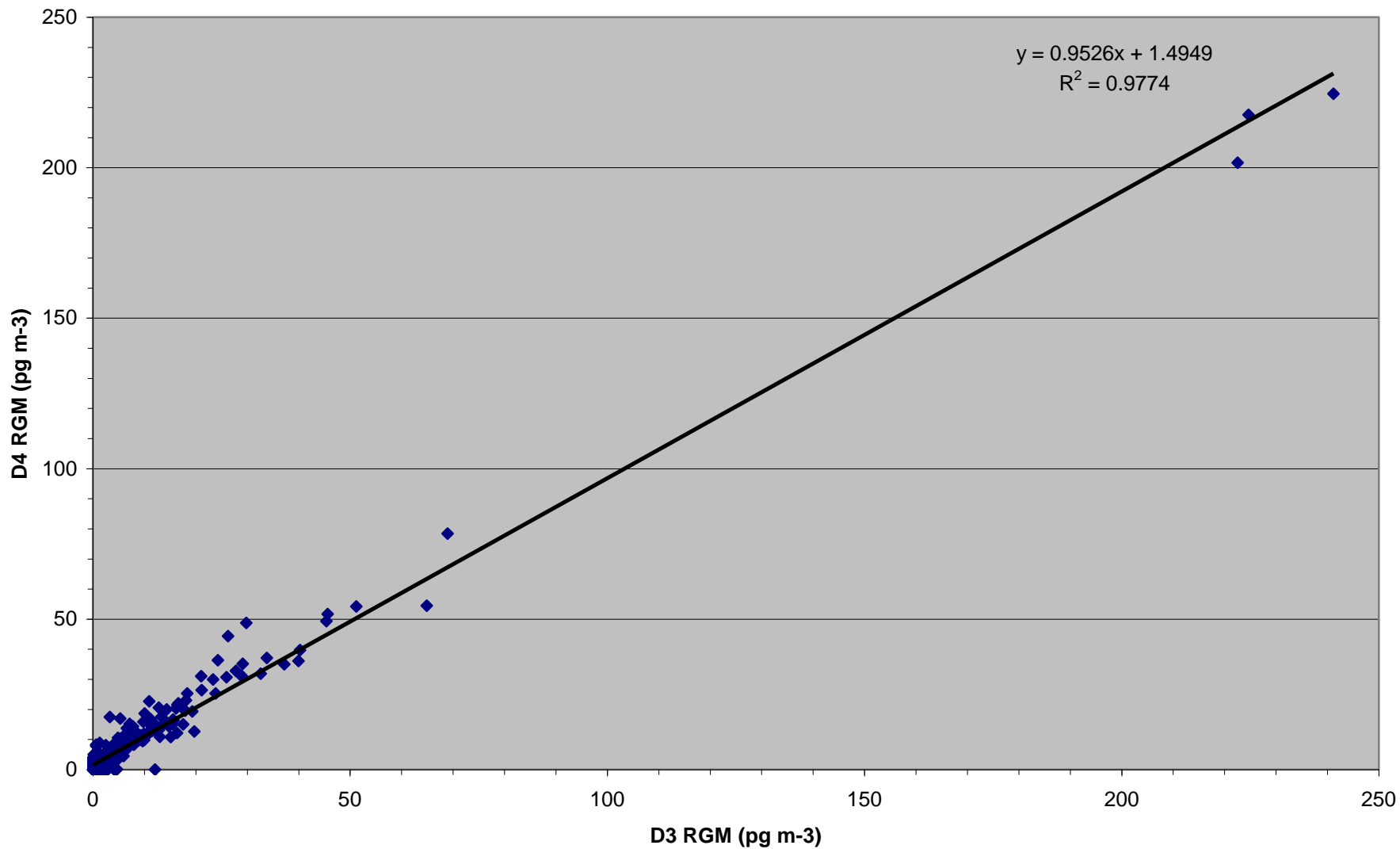
Beltsville site is impacted by a variety of local-regional sources with unique emissions characteristics.

RGM vs SO₂ Summer

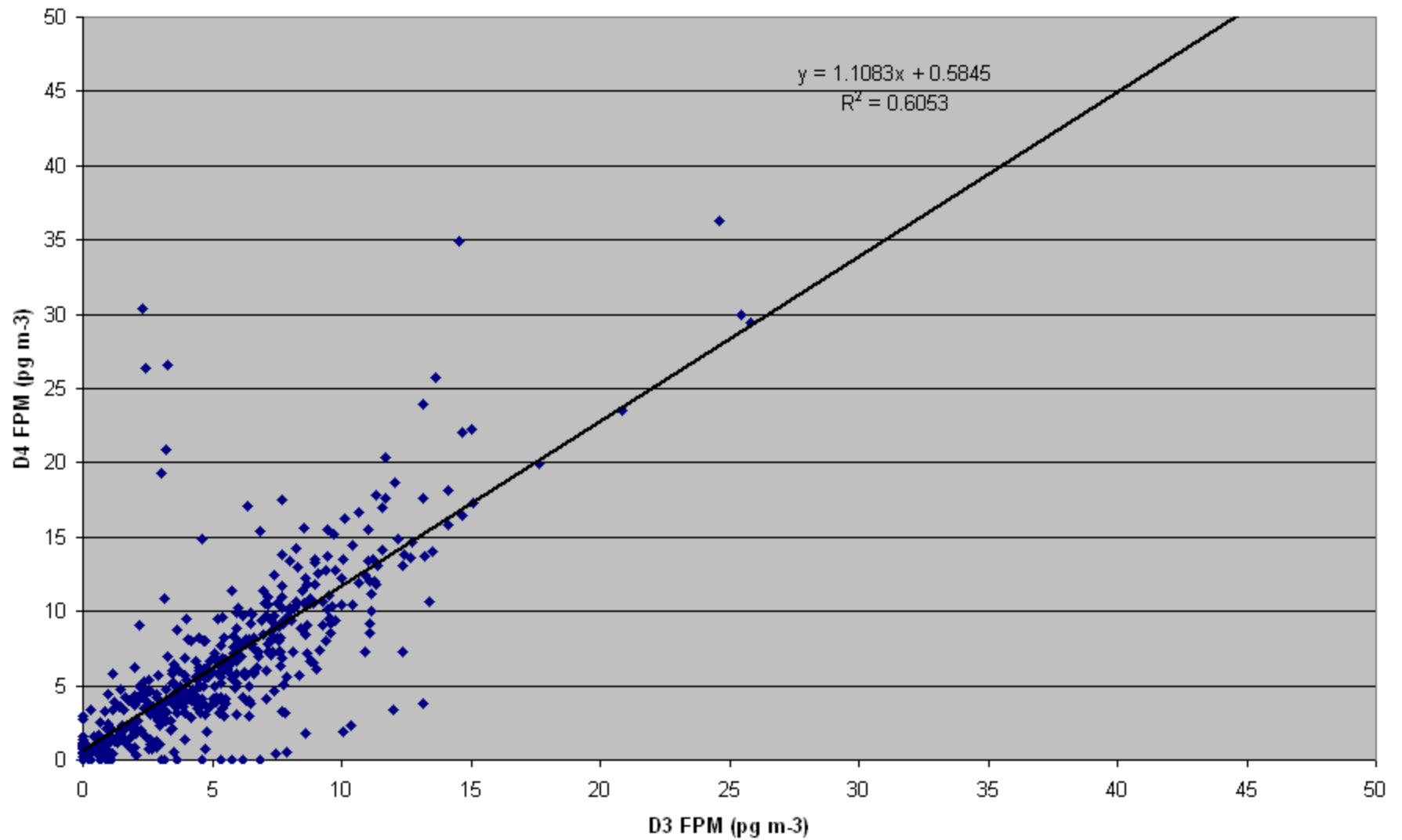


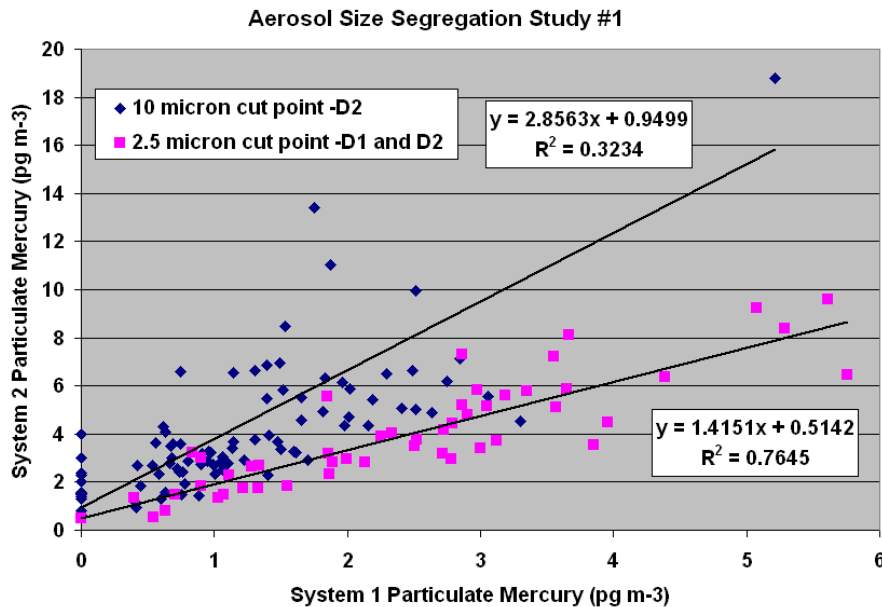
Coupled chemical-meteorological analysis will yield important insights into mercury emissions, transport, transformation, and removal at the site.

D4 vs D3 RGM

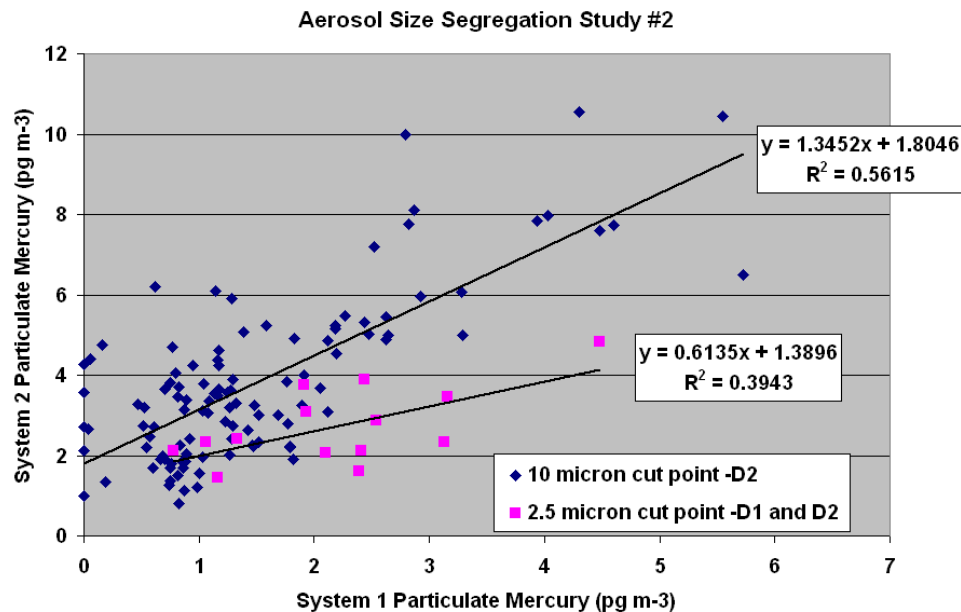


D4 vs D3 FPM





Two systems were configured identically (pink), then System 2 was fitted with a 10-micron cut point elutriator (blue) over the course of several days.



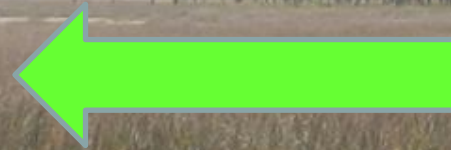
Results suggest that there may be as much mercury in the coarse (sea salt) aerosol fraction as in the fine fraction.

Studies will be repeated periodically at the site.

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Lagrangian Puff Atmospheric Fate and Transport Model

NOAA
HYSPLIT
MODEL

TIME (hours)

0

1

2

...



= mass of pollutant
(changes due to chemical transformations
and deposition that occur at each time step)

Phase partitioning and chemical
transformations of pollutants within the
puff are estimated at each time step

The puff's mass, size,
and location are
continuously tracked...

Initial puff location
is at source, with
mass depending
on emissions rate

Centerline of
puff motion
determined by
wind direction
and velocity

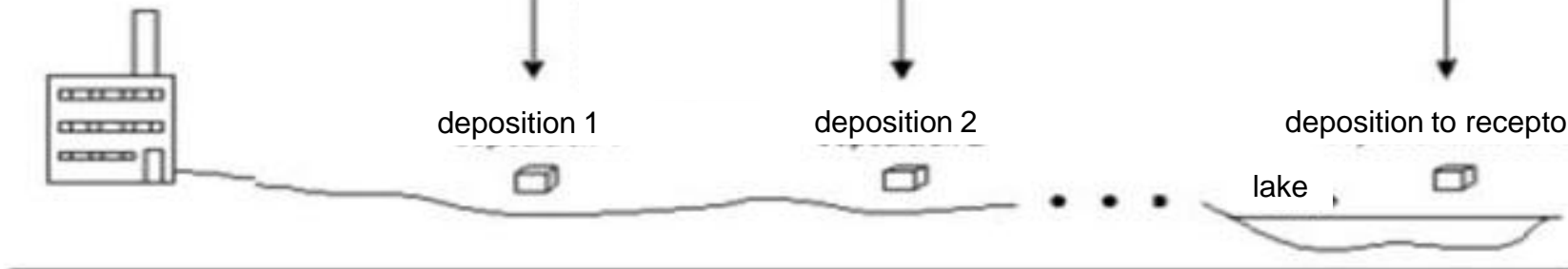
Dry and wet
deposition of the
pollutants
in the puff are
estimated at
each time step.

deposition 1

deposition 2

deposition to receptor

lake



Over the entire modeling period
(e.g., one year), puffs are released
at periodic intervals
(e.g., once every 7 hours).

Each released puff is advected and
dispersed, and the pollutant within
the puff is transformed and deposited.

Release at Time = 1

Release at Time = 8

Release at Time = 15



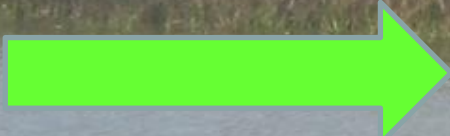
(Evolving) Atmospheric Chemical Reaction Scheme for Mercury

Reaction	Rate	Units	Reference
GAS PHASE REACTIONS			
? $\text{Hg}^0 + \text{O}_3 \rightarrow \text{Hg(p)}$	3.0E-20	$\text{cm}^3/\text{molec-sec}$	Hall (1995)
$\text{Hg}^0 + \text{HCl} \rightarrow \text{HgCl}_2$	1.0E-19	$\text{cm}^3/\text{molec-sec}$	Hall and Bloom (1993)
$\text{Hg}^0 + \text{H}_2\text{O}_2 \rightarrow \text{Hg(p)}$	8.5E-19	$\text{cm}^3/\text{molec-sec}$	Tokos et al. (1998) (upper limit based on experiments)
$\text{Hg}^0 + \text{Cl}_2 \rightarrow \text{HgCl}_2$	4.0E-18	$\text{cm}^3/\text{molec-sec}$	Calhoun and Prestbo (2001)
? $\text{Hg}^0 + \text{OH} \rightarrow \text{Hg(p)}$	8.7E-14	$\text{cm}^3/\text{molec-sec}$	Sommar et al. (2001)
new $\text{Hg}^0 + \text{Br} \rightarrow \text{HgBr}_2$			
AQUEOUS PHASE REACTIONS			
$\text{Hg}^0 + \text{O}_3 \rightarrow \text{Hg}^{+2}$	4.7E+7	$(\text{molar-sec})^{-1}$	Munthe (1992)
$\text{Hg}^0 + \text{OH} \rightarrow \text{Hg}^{+2}$	2.0E+9	$(\text{molar-sec})^{-1}$	Lin and Pehkonen(1997)
$\text{HgSO}_3 \rightarrow \text{Hg}^0$	$T^*e^{((31.971^*T)-12595.0)/T}$ sec ⁻¹ [T = temperature (K)]		Van Loon et al. (2002)
? $\text{Hg(II)} + \text{HO}_2 \rightarrow \text{Hg}^0$	~ 0	$(\text{molar-sec})^{-1}$	Gardfeldt & Jonnson (2003)
$\text{Hg}^0 + \text{HOCl} \rightarrow \text{Hg}^{+2}$	2.1E+6	$(\text{molar-sec})^{-1}$	Lin and Pehkonen(1998)
$\text{Hg}^0 + \text{OCl}^{-1} \rightarrow \text{Hg}^{+2}$	2.0E+6	$(\text{molar-sec})^{-1}$	Lin and Pehkonen(1998)
$\text{Hg(II)} \leftrightarrow \text{Hg(II)}_{(\text{soot})}$	9.0E+2	liters/gram; t = 1/hour	eqbrm: Seigneur et al. (1998) rate: Bullock & Brehme (2002).
$\text{Hg}^{+2} + h\nu \rightarrow \text{Hg}^0$	6.0E-7	$(\text{sec})^{-1}$ (maximum)	Xiao et al. (1994); Bullock and Brehme (2002)

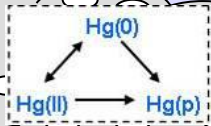
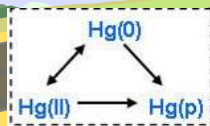
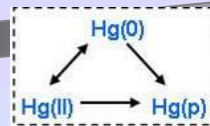
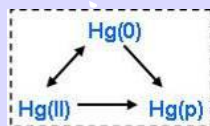
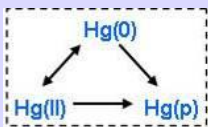
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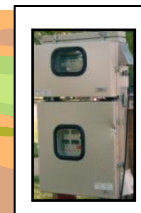
**Hg from
other sources:
local, regional
& more distant**



**Measurement
of wet
deposition**

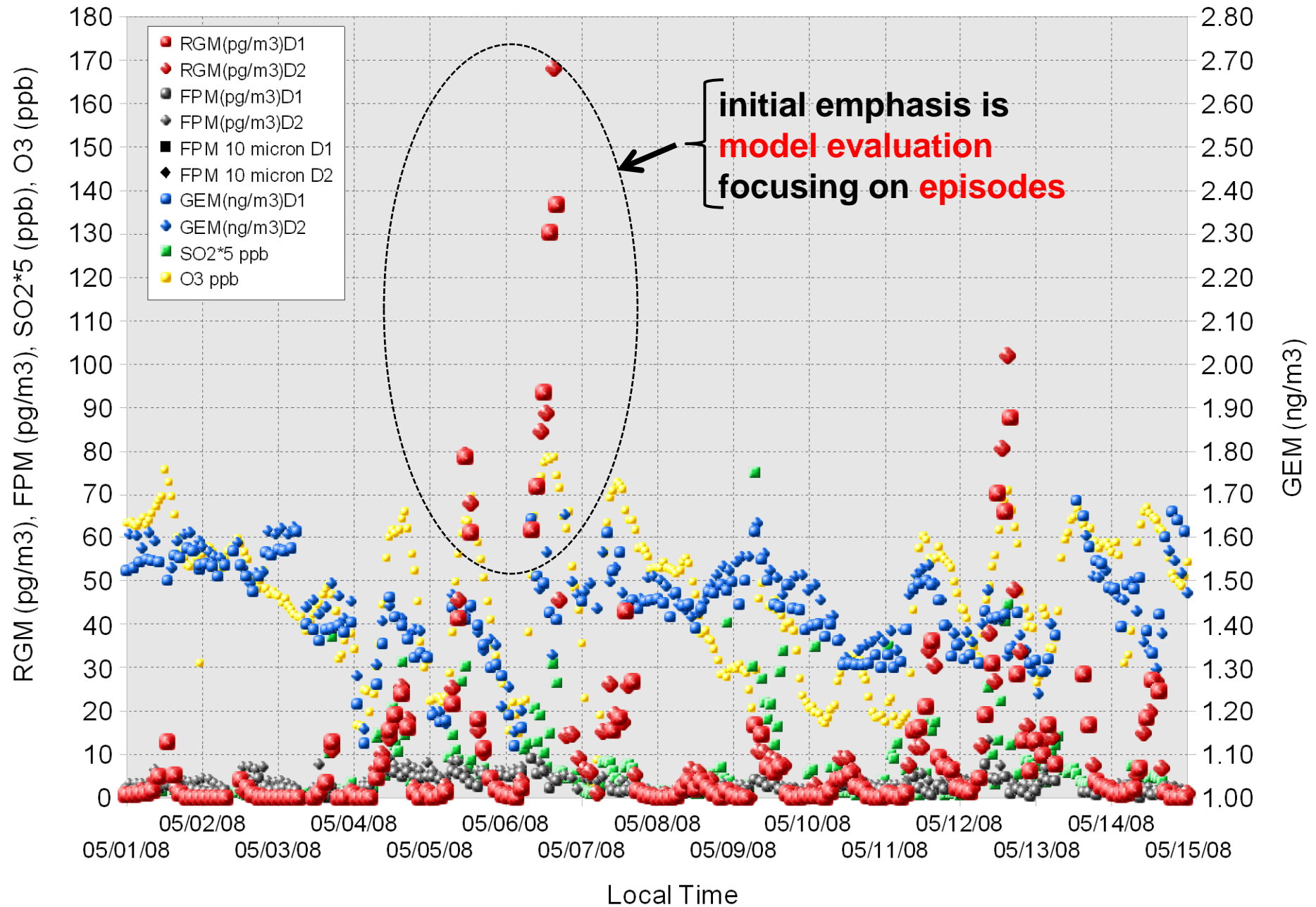


**Measurement
of ambient air
concentrations**



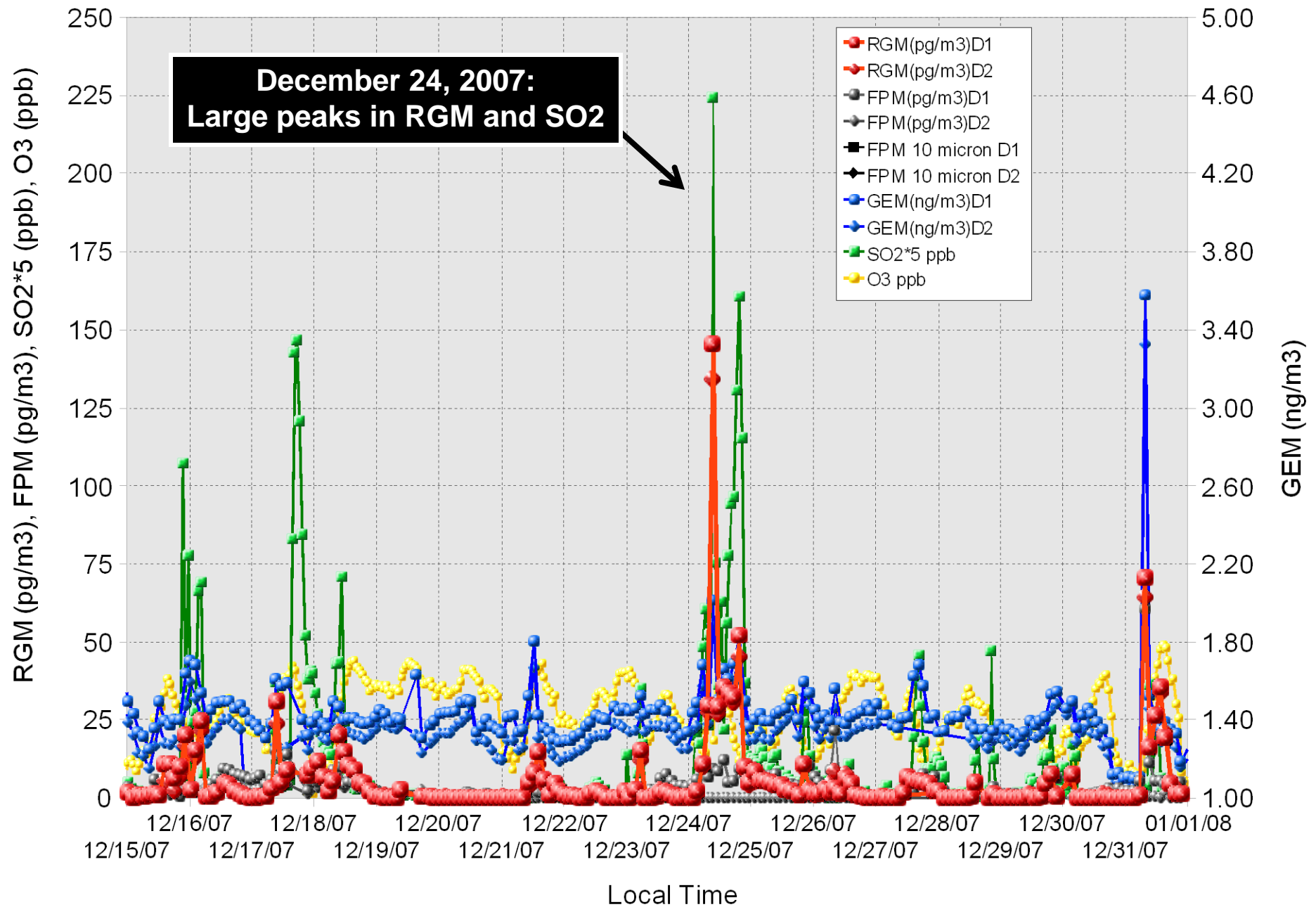
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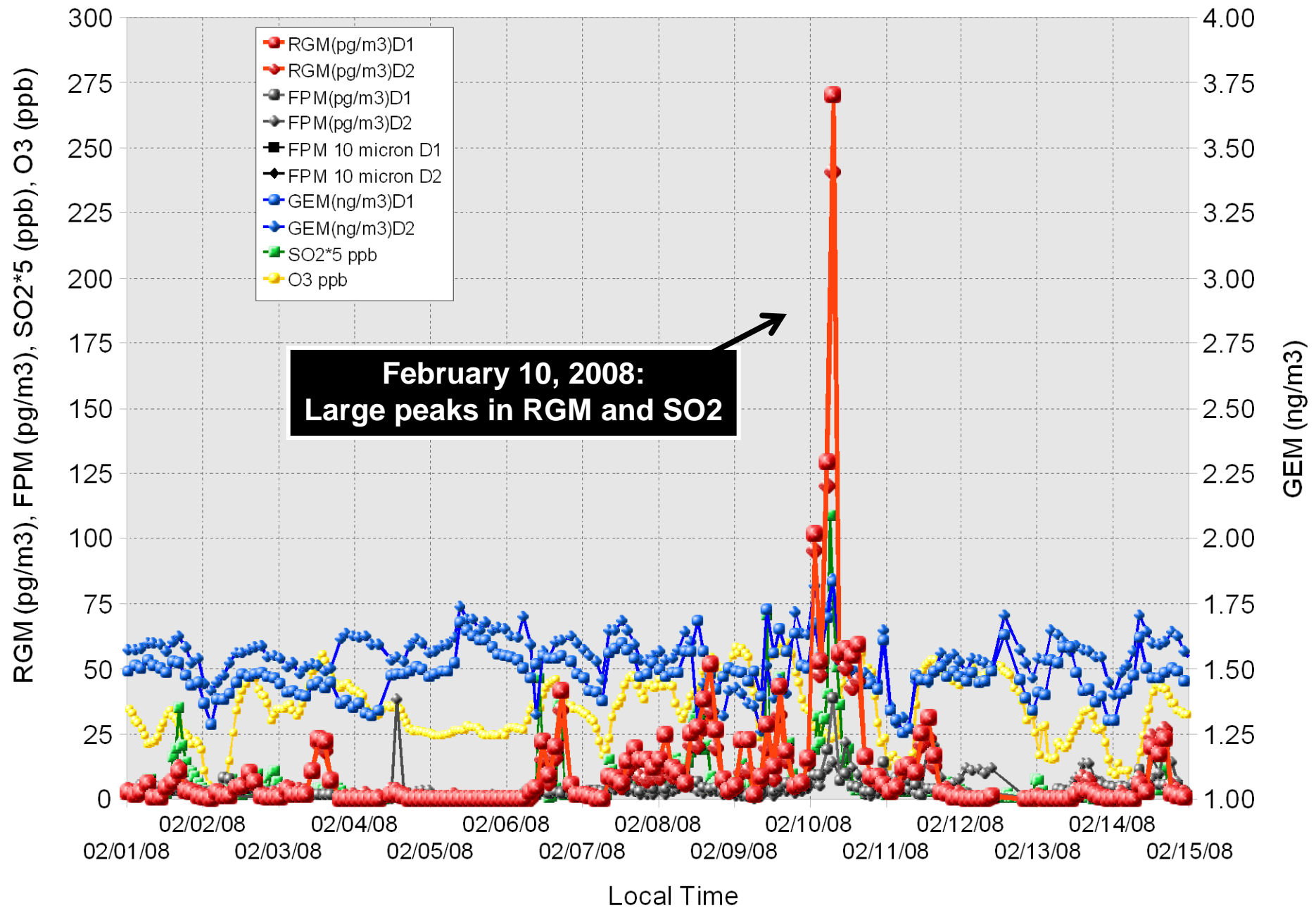
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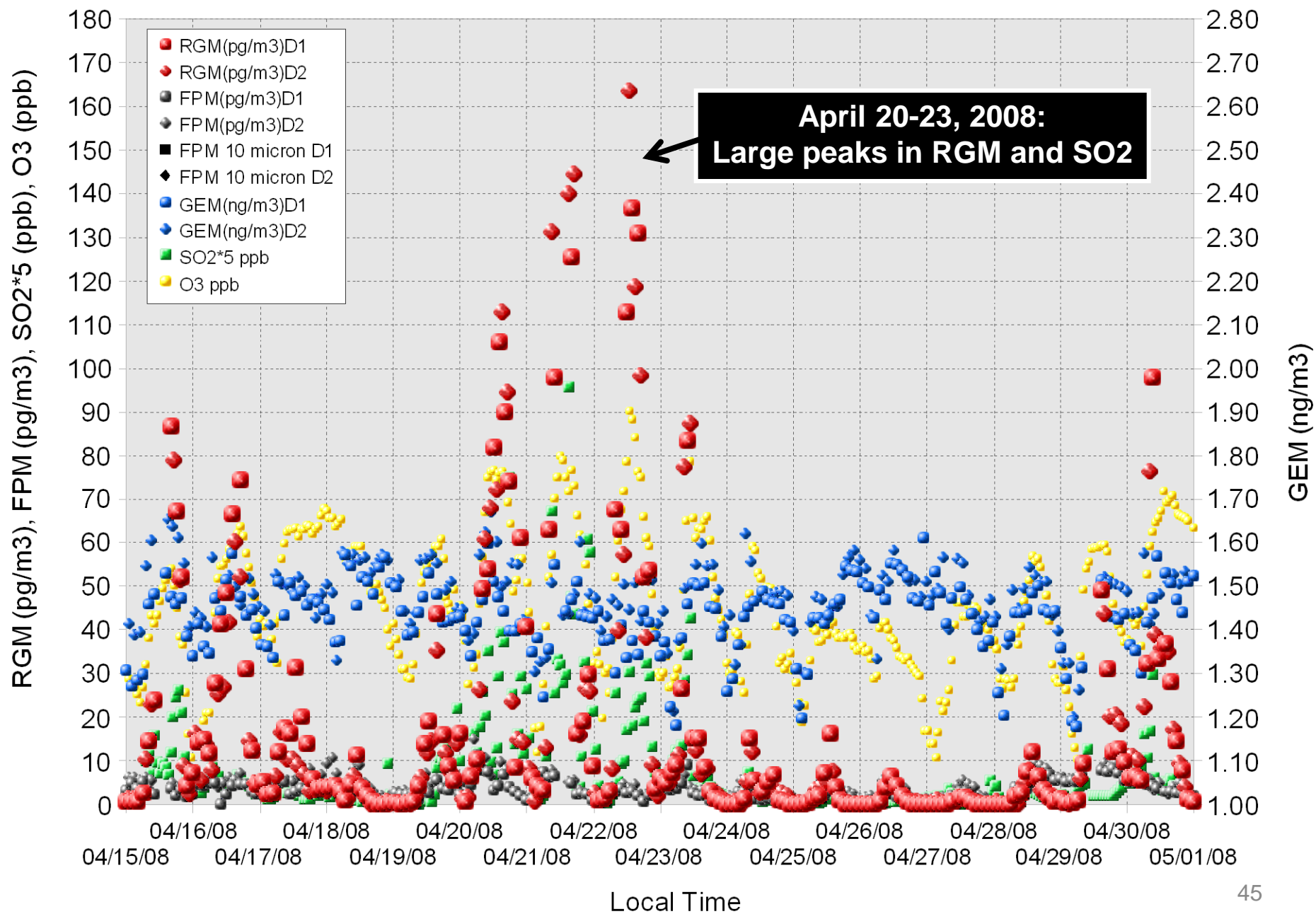
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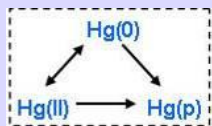
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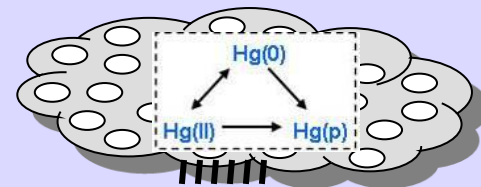
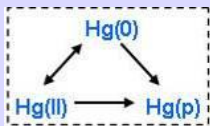
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**Hg from
other sources:
local, regional
& more distant**



**atmospheric
deposition
to the water
surface**



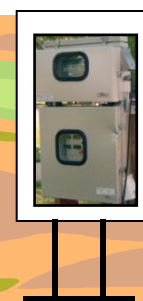
**atmospheric
deposition
to the
watershed**



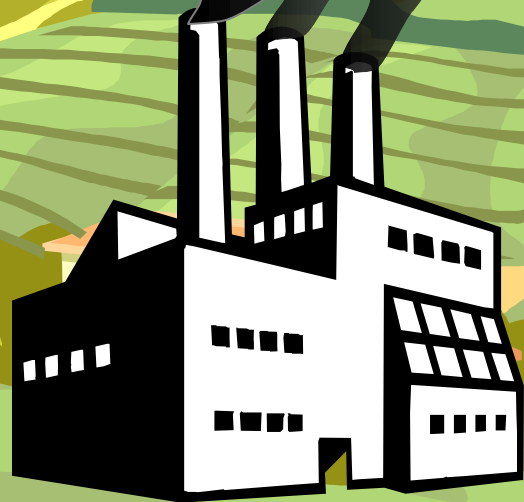
**Measurement
of wet
deposition**



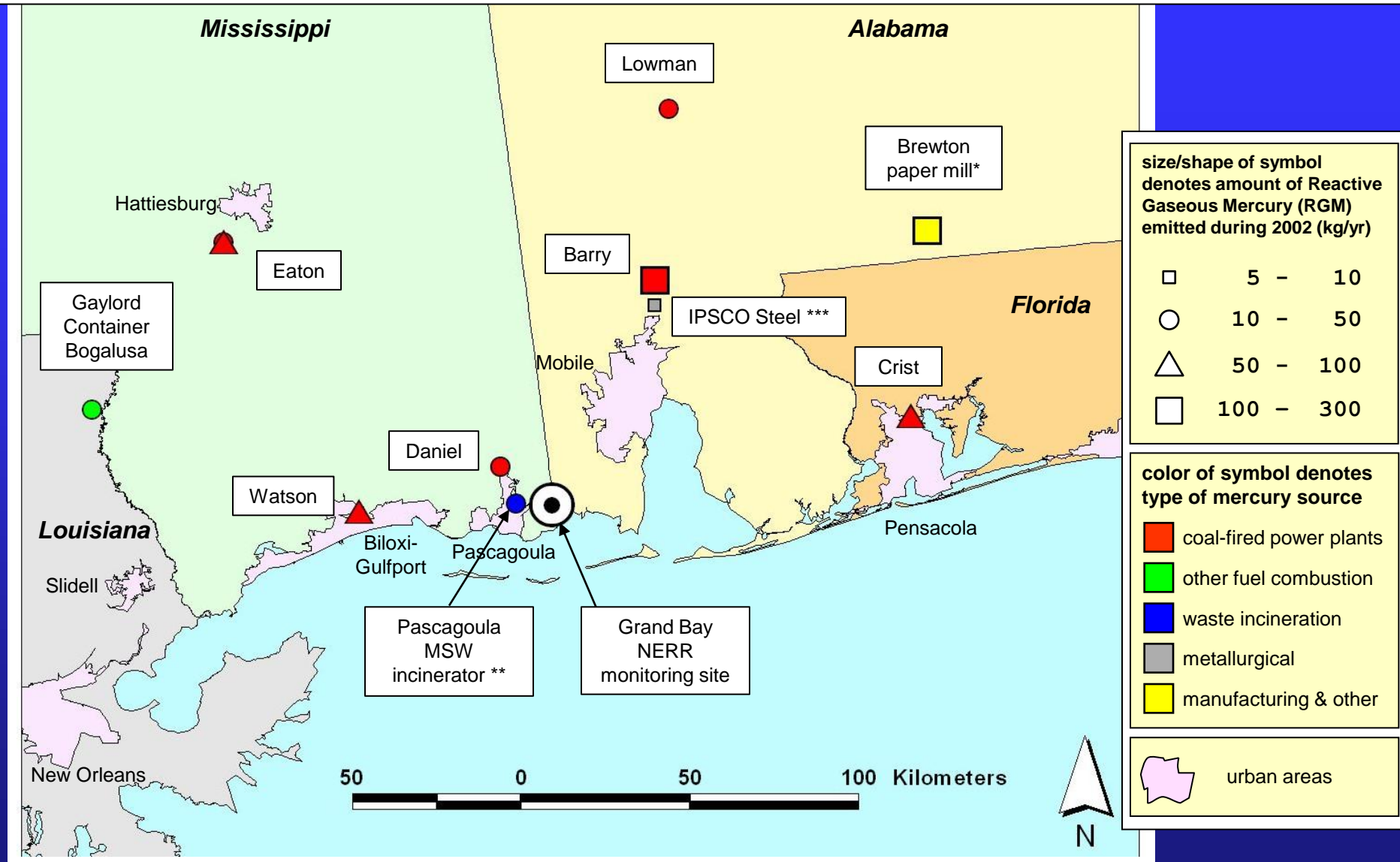
**Measurement
of ambient air
concentrations**



**An essential factor in carrying
out a meaningful model
evaluation in cases where
local/regional sources may be
important is to have accurate
emissions data for
local/regional sources, valid
for the time of the episode
being studied**



Grand Bay NERR sampling site, with large point sources of Reactive Gaseous Mercury in the region, based on the EPA's 2002 National Emissions Inventory

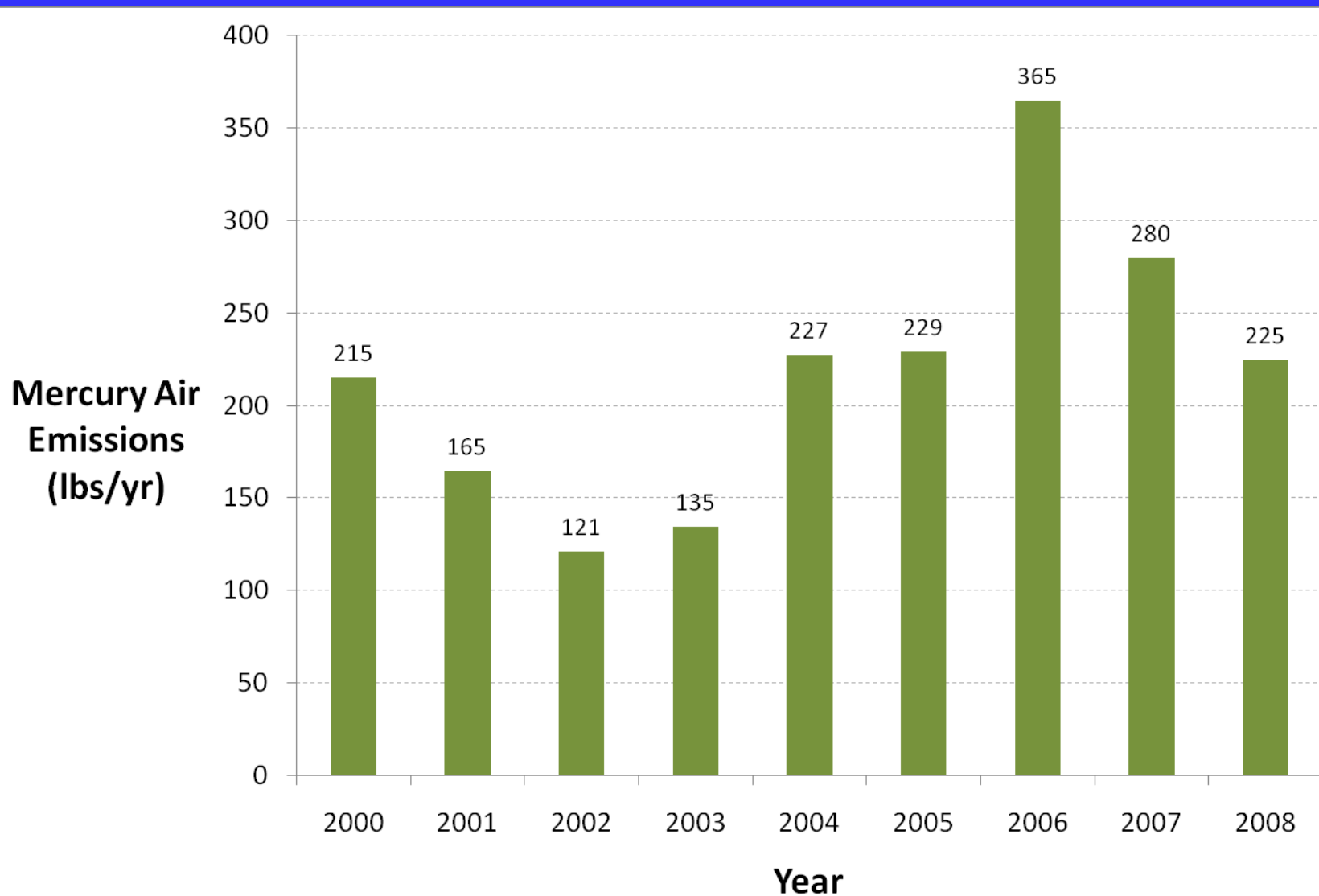


* Brewton paper mill mercury emissions included in 2002 NEI , but do not appear to be in 2000-2008 TRI

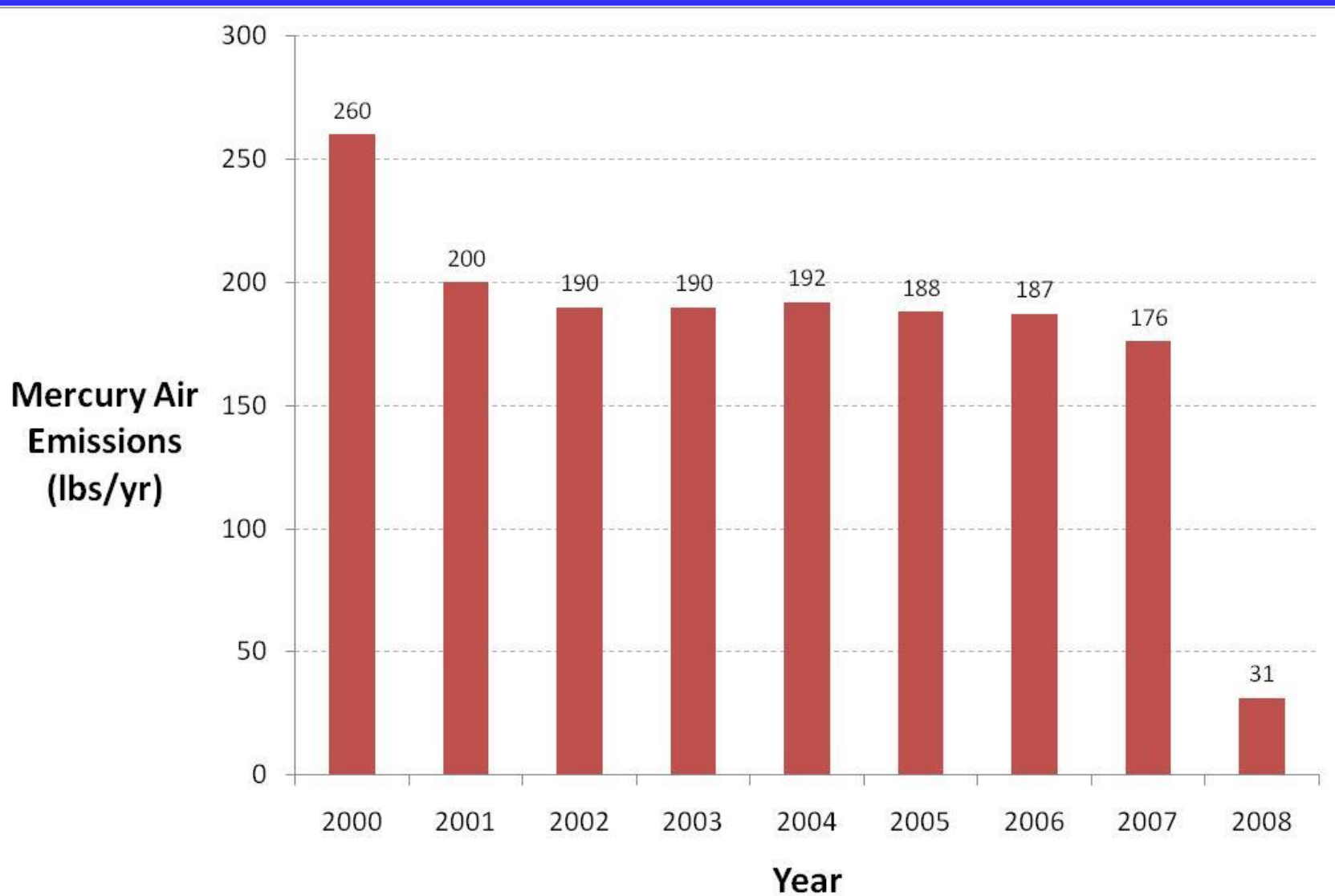
** Pascagoula MSW incinerator mercury emissions included in 2002 NEI but incineration ceased in Jan 2001

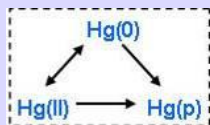
*** Ipsco Steel had significant mercury emissions in 2002 NEI, but negligible emissions reported in 2008 TRI

Mercury Air Emissions from the Victor J. Daniel Power Plant as reported to the Toxic Release Inventory

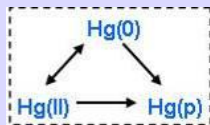


Mercury Air Emissions from Charles R. Lowman Power Plant as reported to the Toxic Release Inventory



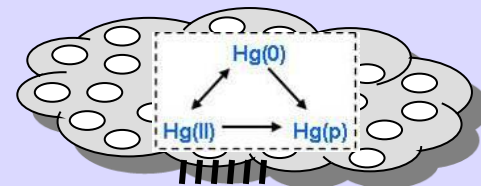
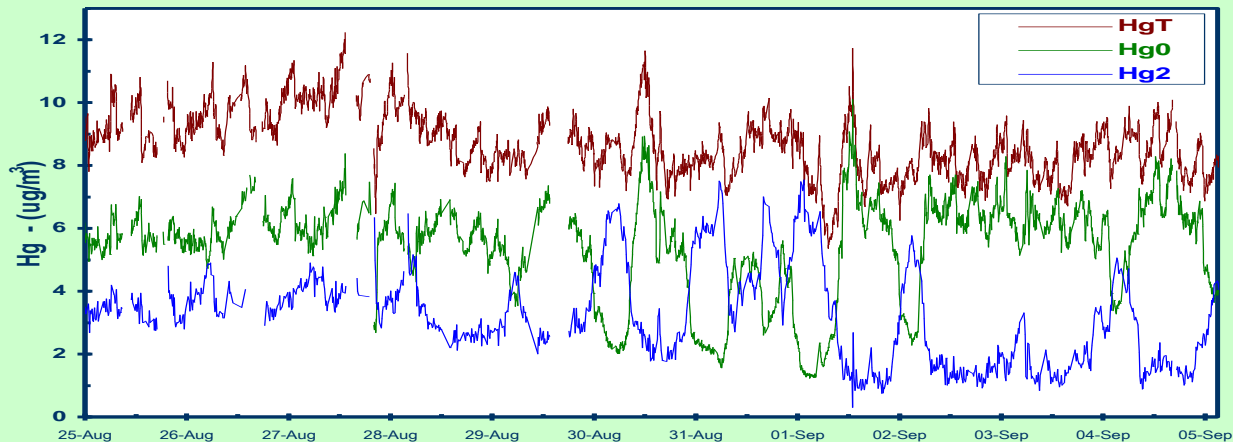


**Hg from
other sources:
local, regional
& more distant**



Series 3300 CEM - Continuous Speciated Mercury Data

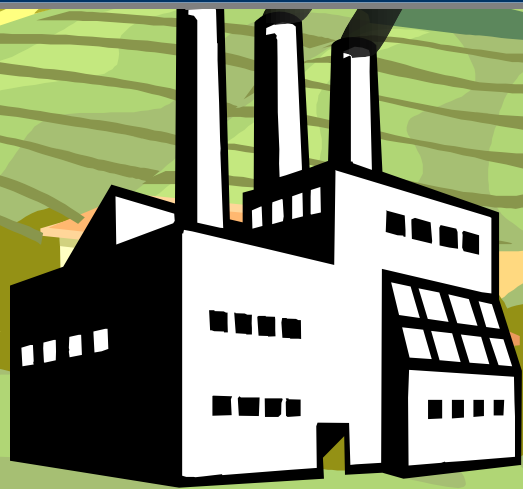
Resolution: 2.5 min Duration: 11 Days

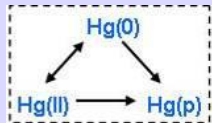


atmospheric
deposition
to the water
surface

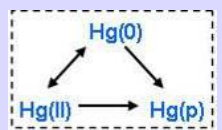
Measurement
of wet
deposition

Measurement
of ambient air
concentrations

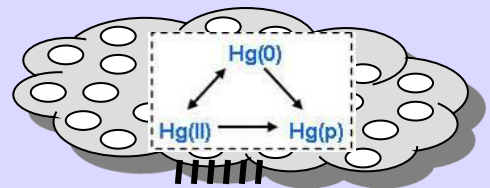
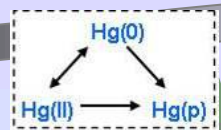
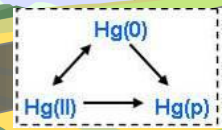




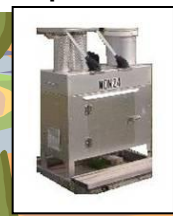
Hg from
other sources:
local, regional
& more distant



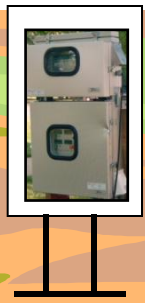
Another critical factor in carrying out a meaningful model evaluation in cases where local/regional sources may be important is to have accurate meteorological data to drive the model



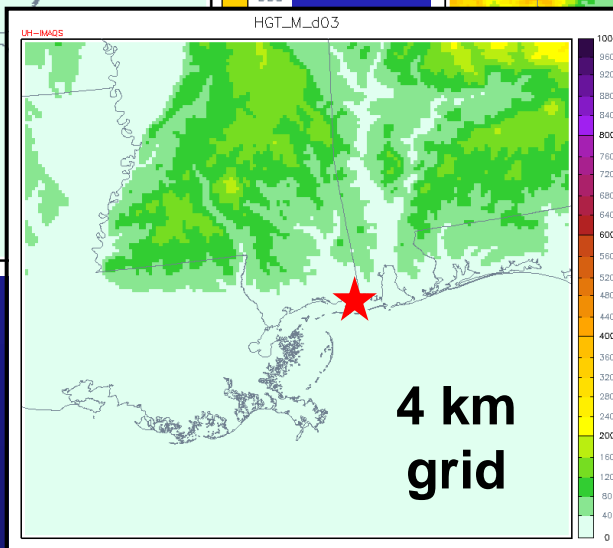
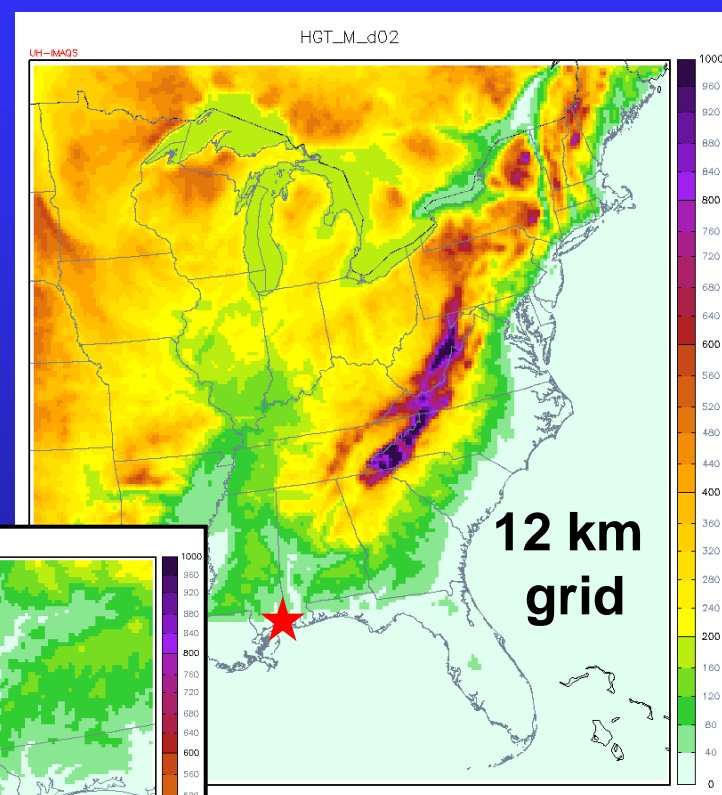
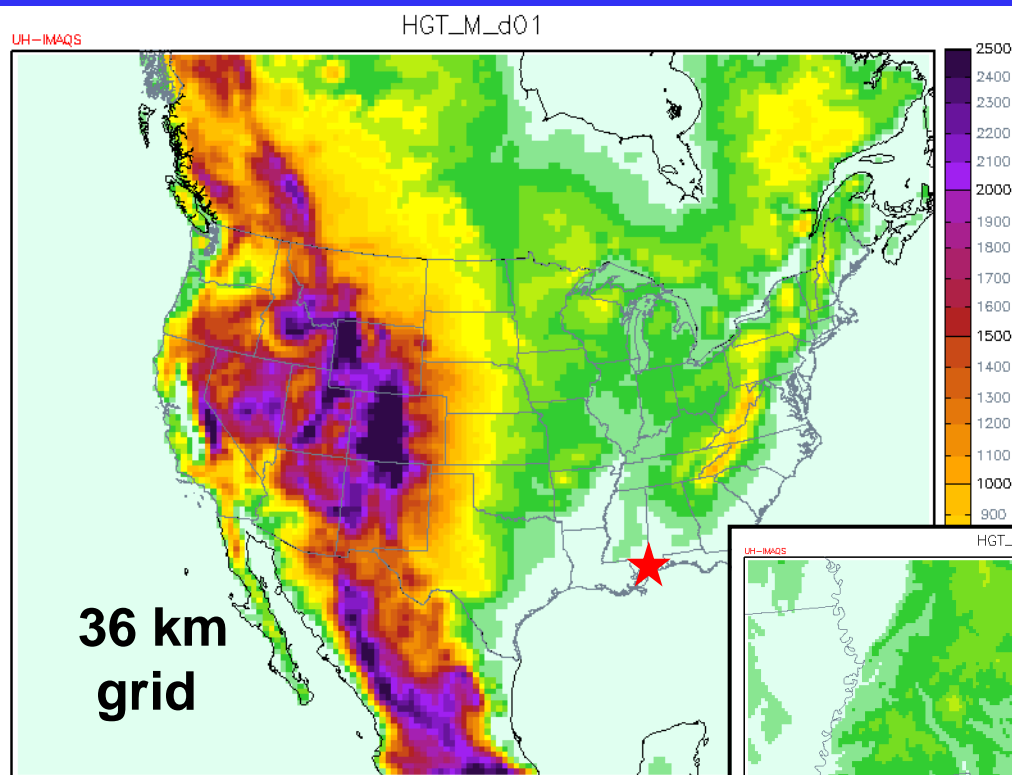
Measurement
of wet
deposition



Measurement
of ambient air
concentrations



High-resolution meteorological simulations being carried out for episodes at the Grand Bay NERR [★] by Dr. Fantine Ngan, a post-doc at NOAA ARL, and independently by Dr. Rao Dodla of Jackson State University



*Terrain height
of 3 domains*

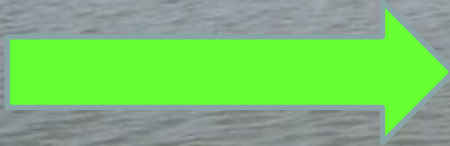
Fantine Ngan, NOAA ARL

1. Measurements

- A. Site Locations and Settings
- B. Current Suite of Measurements
- C. Intensive (this Summer)
- D. Data – some examples

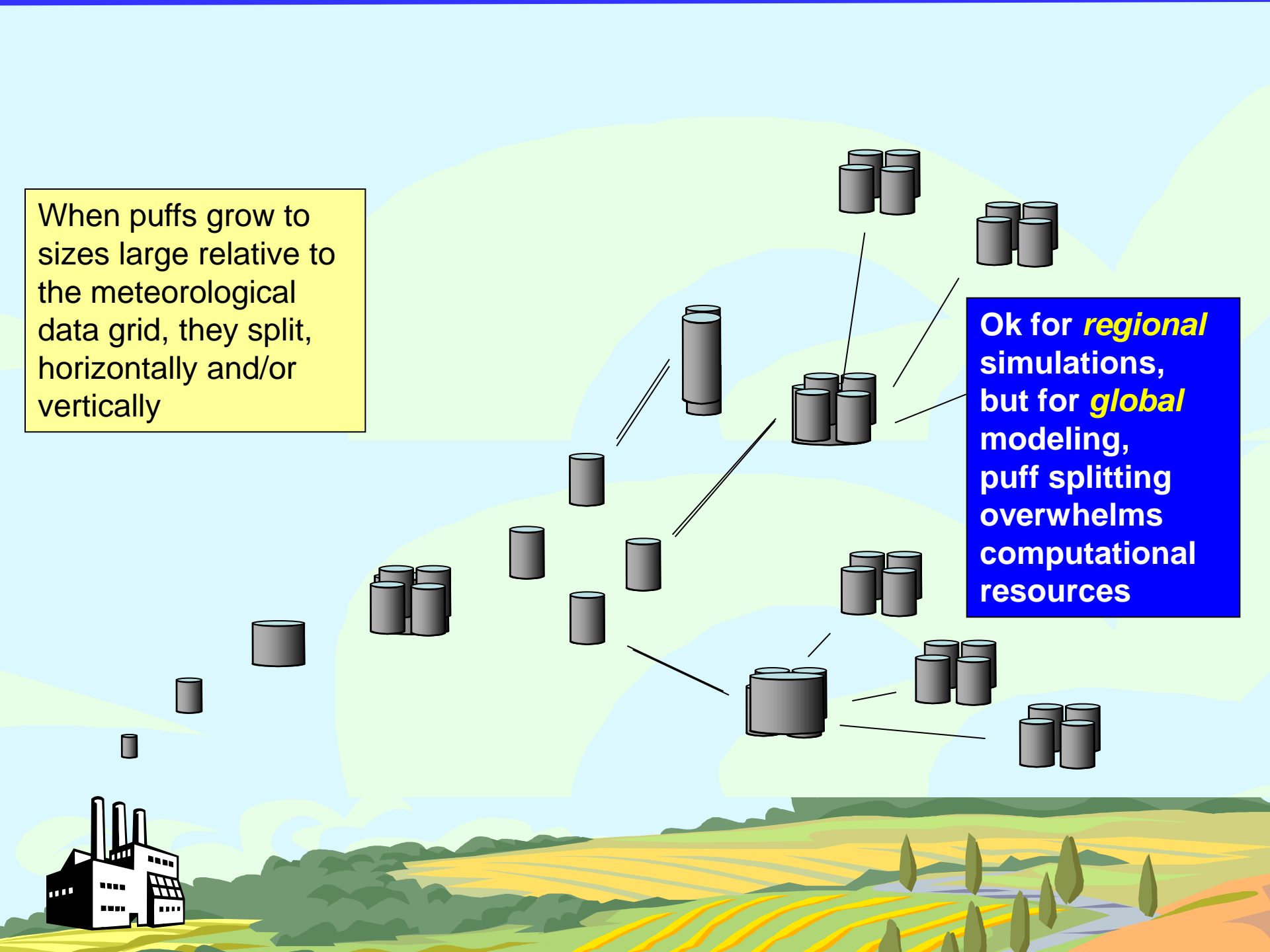
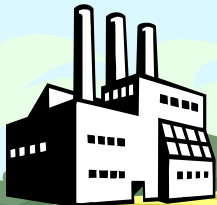
2. Modeling

- A. Episodes for Model Evaluation
- B. Emissions and Met Data -- Fine-Scale
- C. Evolution of the HYSPLIT-Hg Model
- D. Upcoming Great Lakes Project



When puffs grow to sizes large relative to the meteorological data grid, they split, horizontally and/or vertically

Ok for *regional* simulations, but for *global* modeling, puff splitting overwhelms computational resources

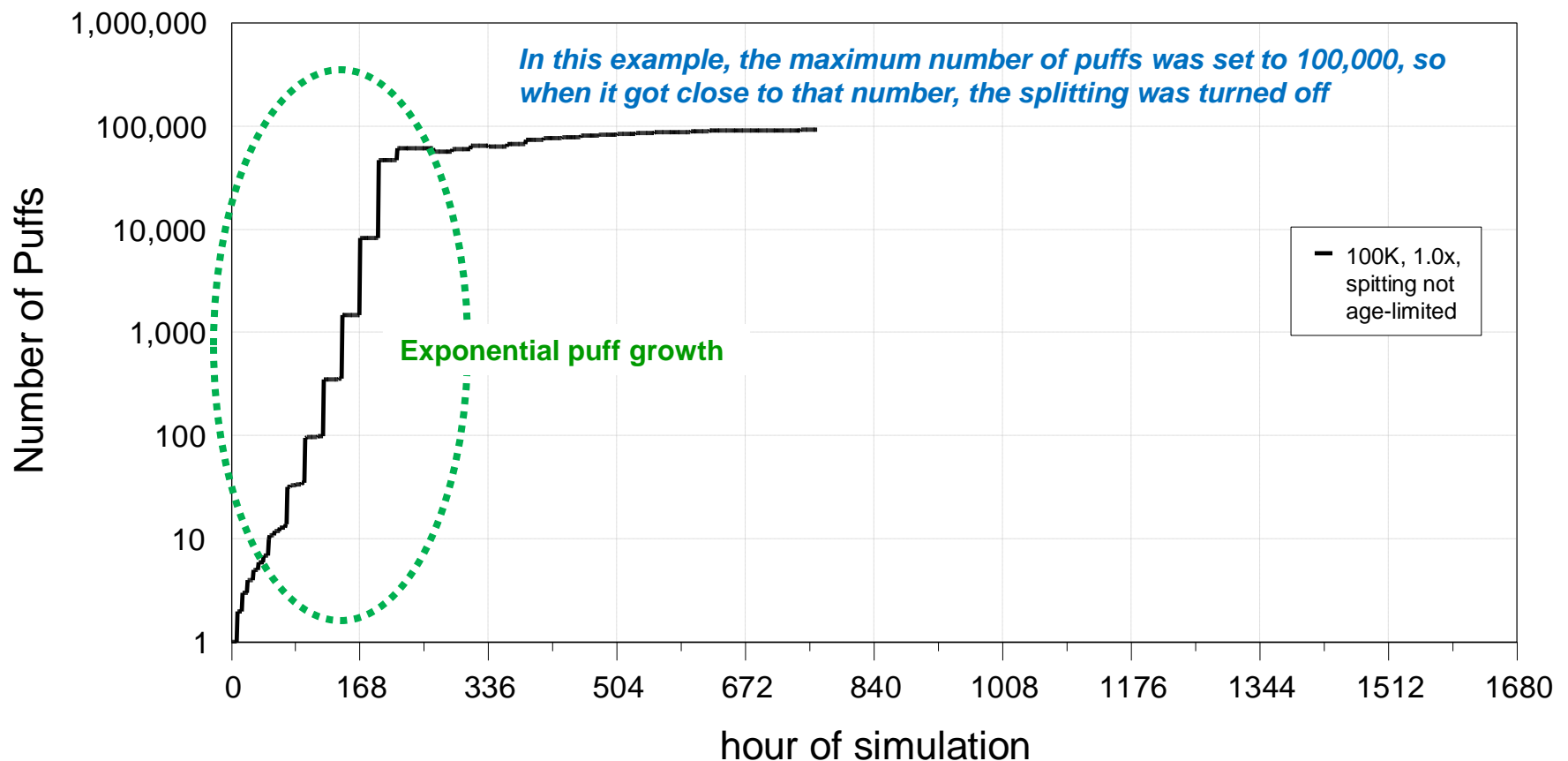


Due to puff splitting, the number of puffs quickly overwhelms numerical resources

Evolution of Number of Puffs

as a function of MAXPAR and merge parameter multiplication factor

elem emit; growth not stopped; splitting not age-limited; source at lat = 30, long = 105 (China)

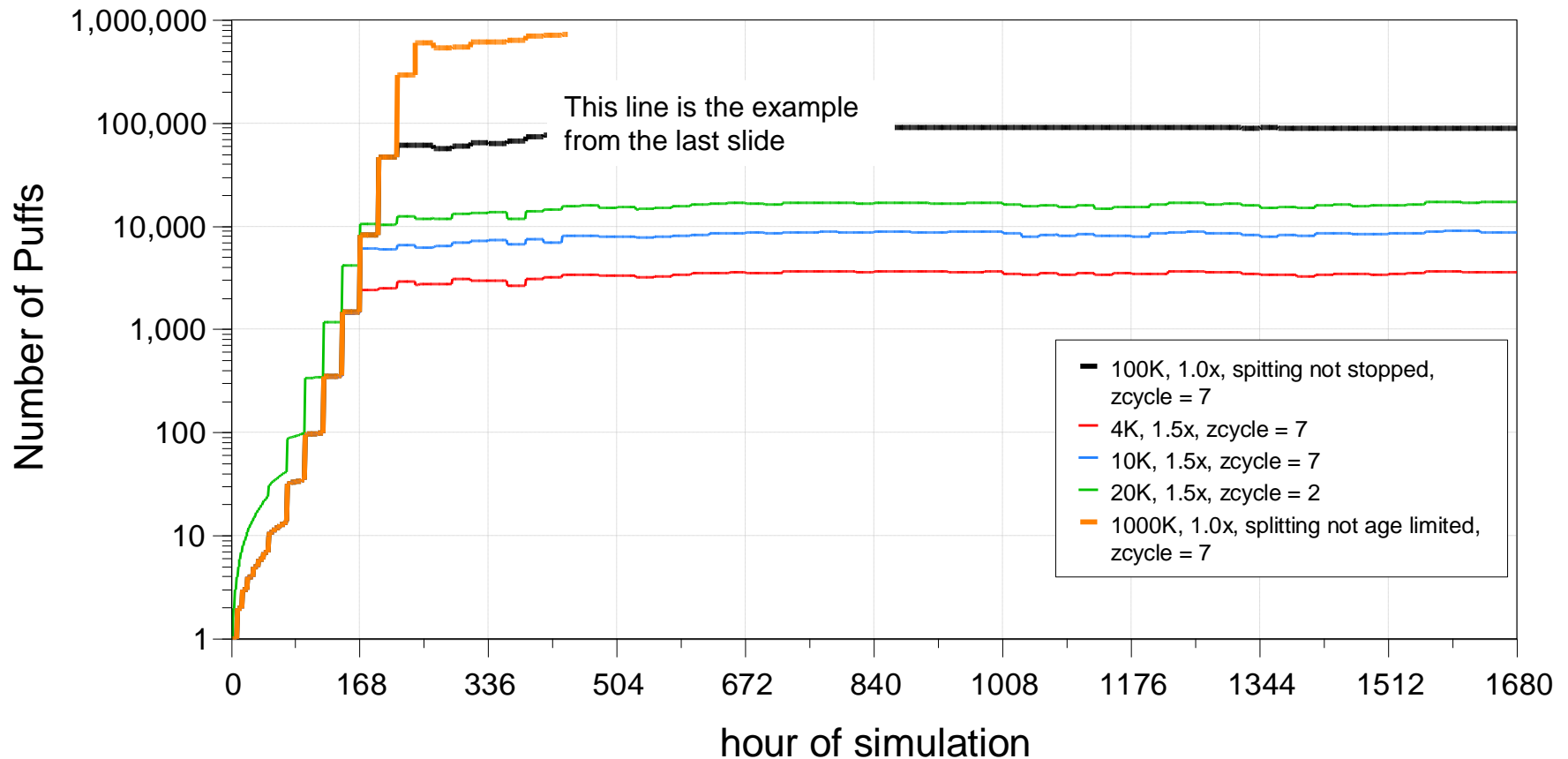


In each test, the number of puffs rises to the maximum allowable within ~ one week (and then splitting stops...)

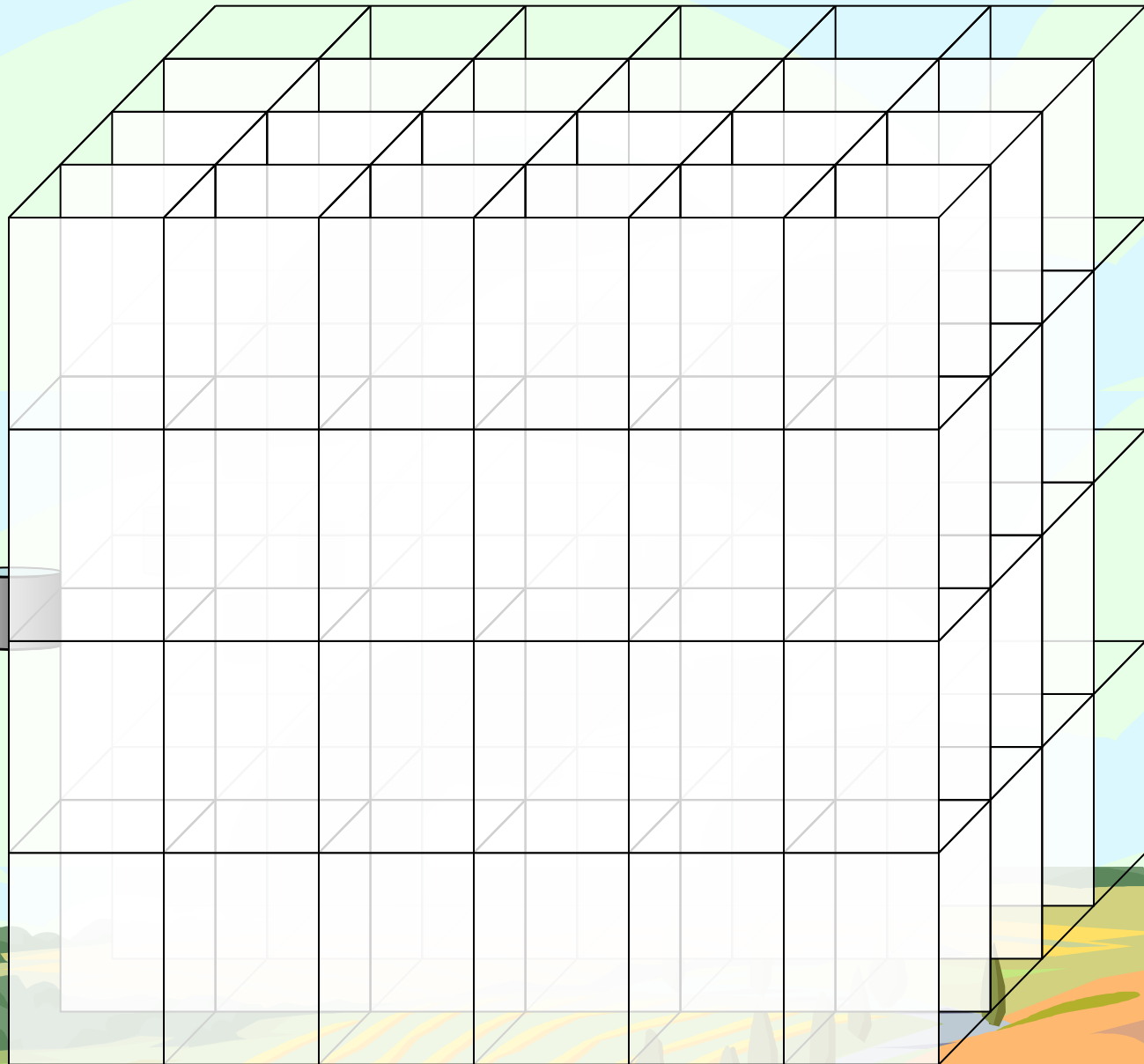
Evolution of Number of Puffs

as a function of MAXPAR and merge parameter multiplication factor

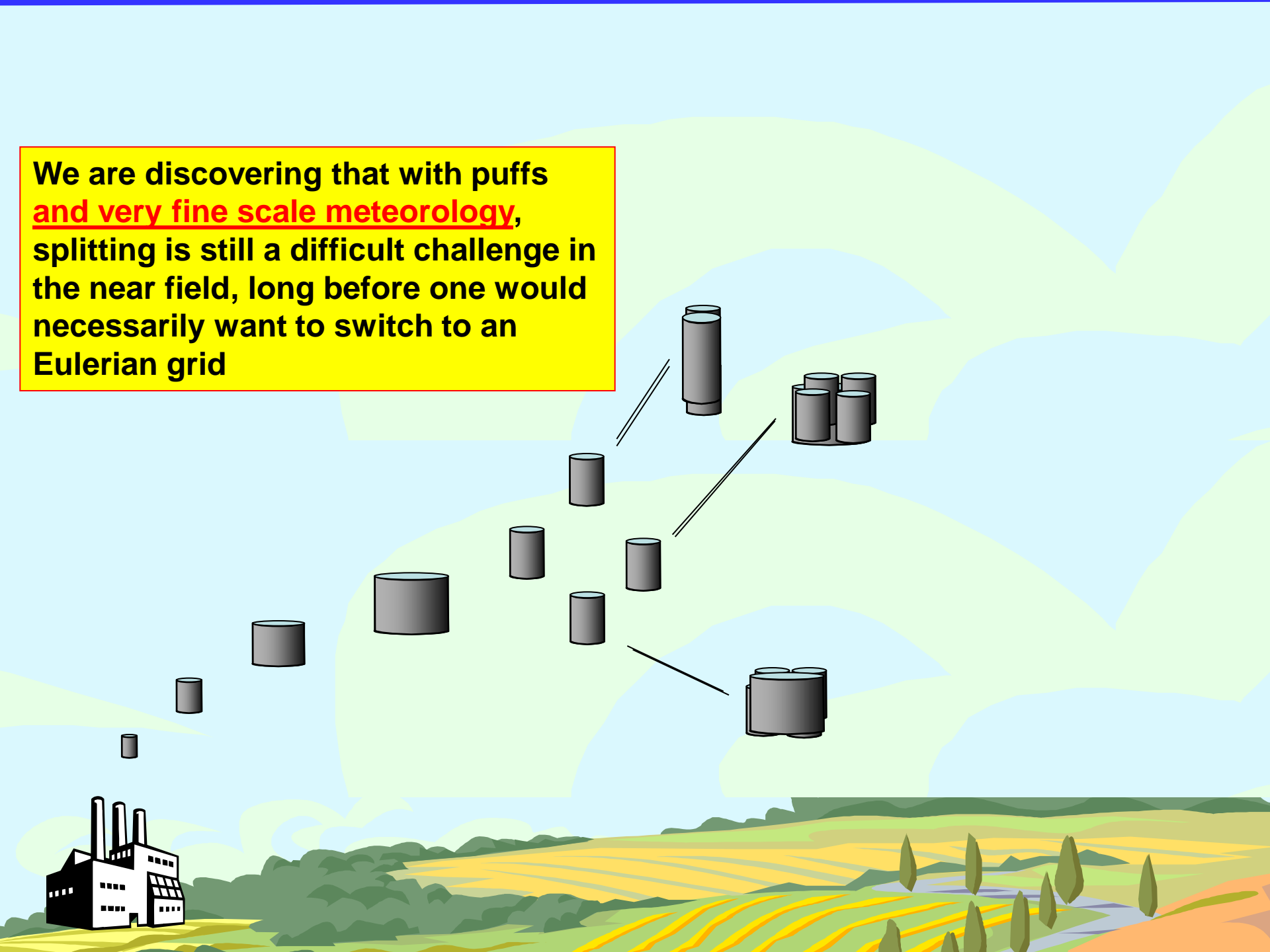
elem emit; growth not stopped; splitting stopped after 168 hours; source at lat = 30, long = 105 (China)



In the new version of HYSPLIT (4.9), puffs are “dumped” into an Eulerian grid after a specified time (e.g., 96 hrs), and the mercury is simulated on that grid from then on...

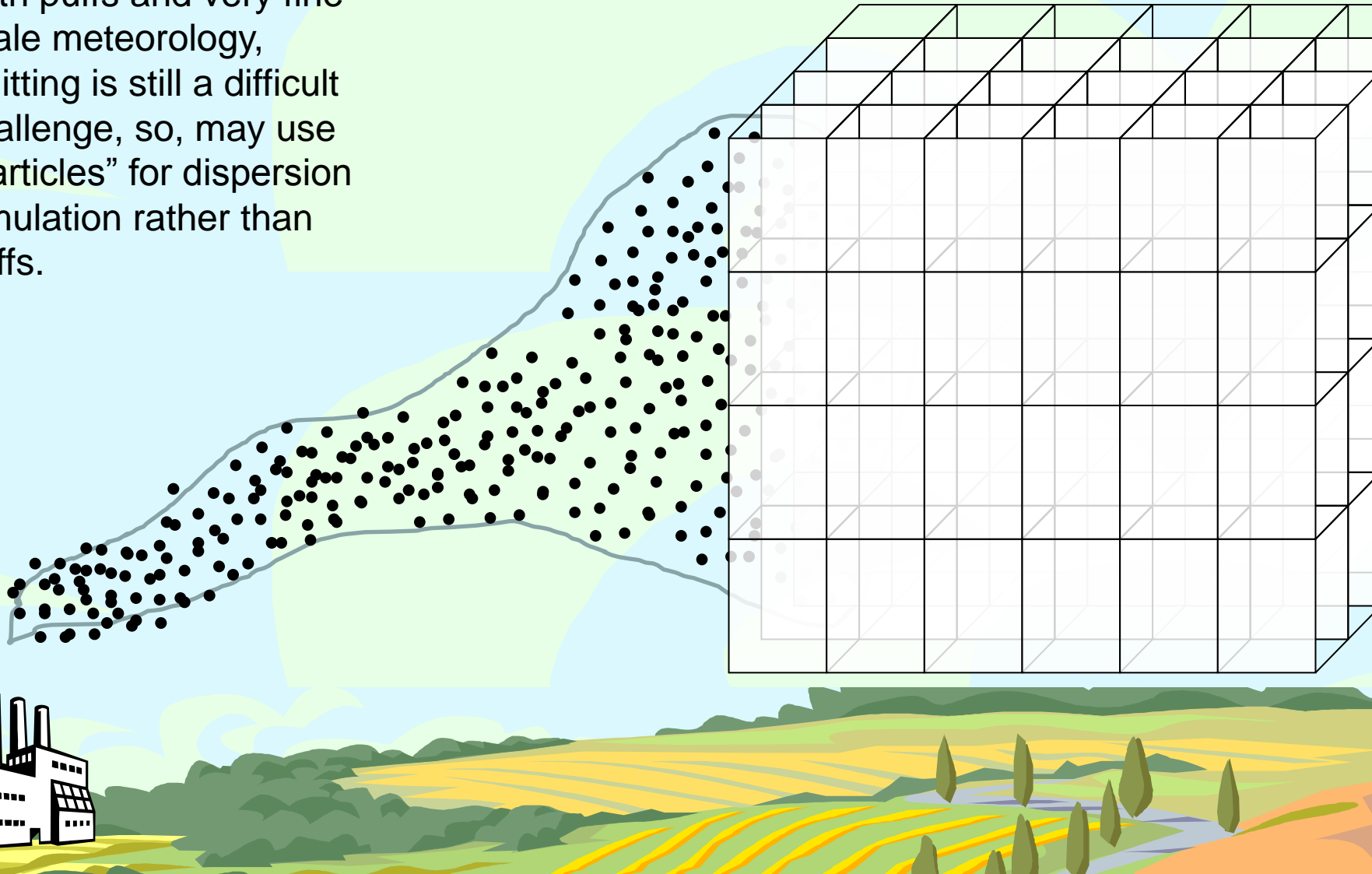


We are discovering that with puffs
and very fine scale meteorology,
splitting is still a difficult challenge in
the near field, long before one would
necessarily want to switch to an
Eulerian grid



With puffs and very fine scale meteorology, splitting is still a difficult challenge, so, may use “particles” for dispersion simulation rather than puffs.

The particles would still be transferred to an Eulerian grid after a given time

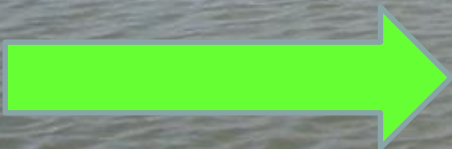


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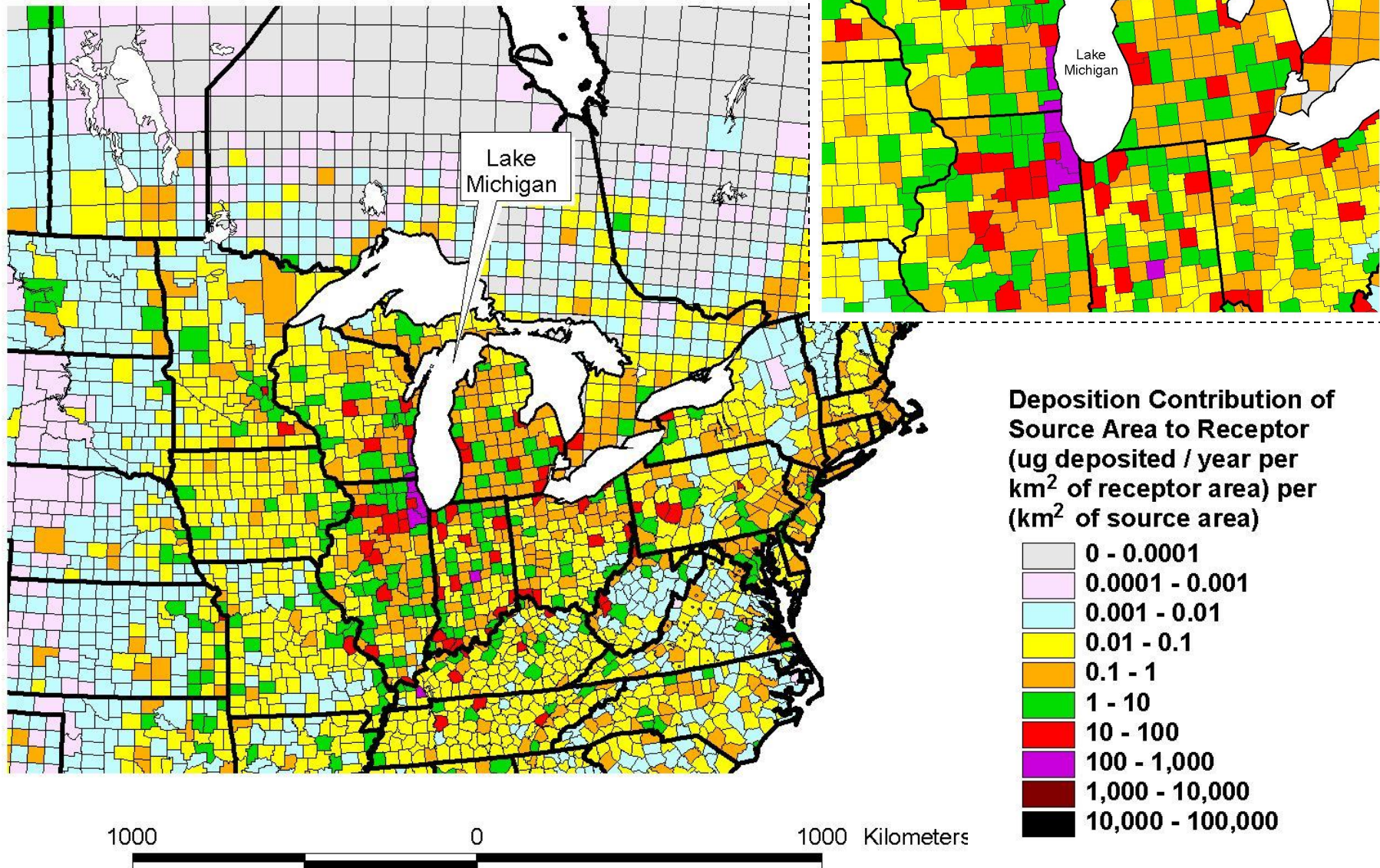


Have done a lot of source-receptor modeling work for the Great Lakes.

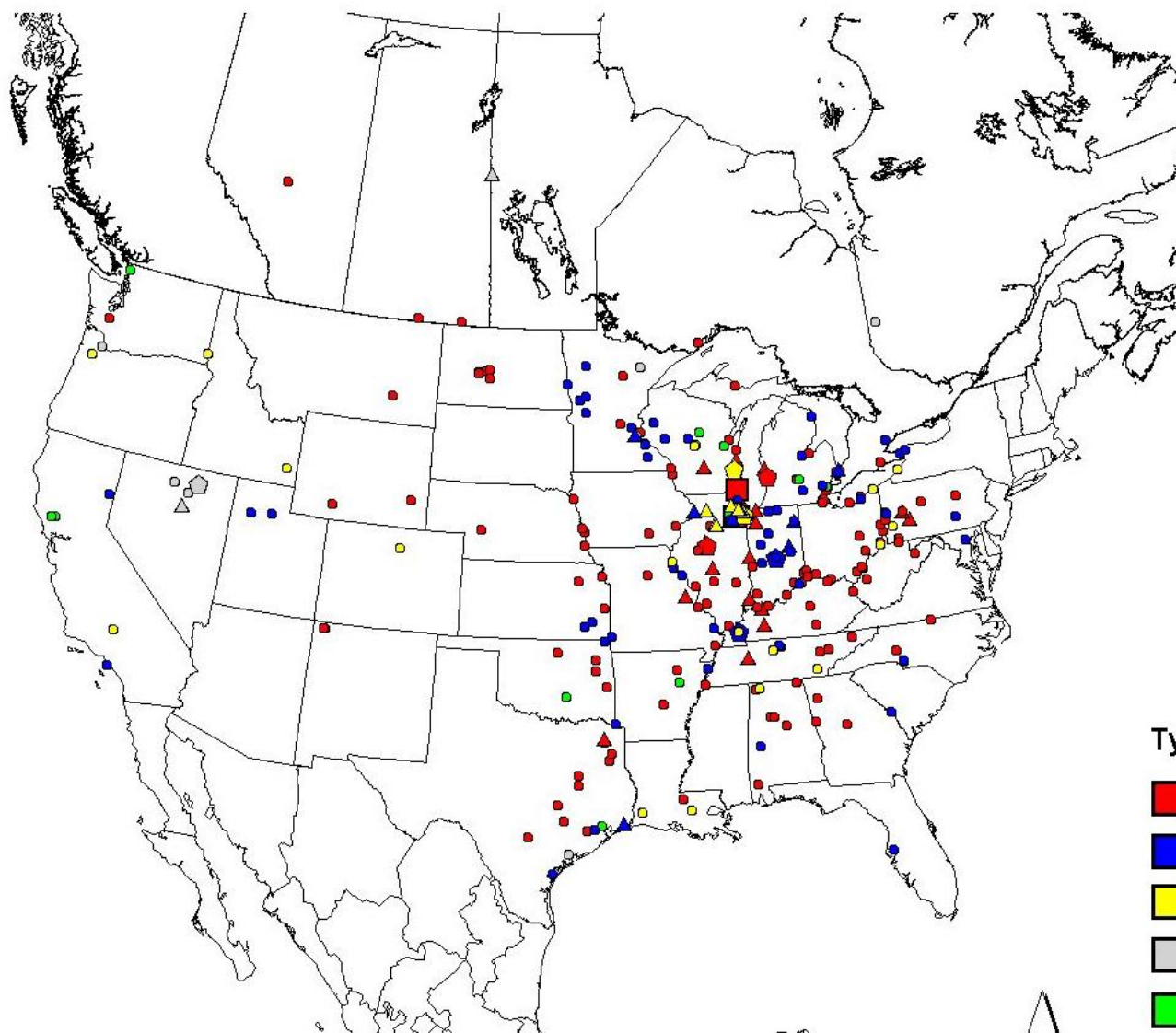
New, expanded study of source attribution for atmospheric mercury deposition to the Great Lakes, as part of the Great Lakes Restoration Initiative



Geographical Distribution of 1999 Direct Deposition Contributions to Lake Michigan



Largest atmospheric deposition contributors to Lake Michigan based on 1999-2000 emissions



Fraction of total modeled deposition contributed by a particular source

- 0.1 - 0.3 %
- △ 0.3 - 1 %
- ⬠ 1 - 3 %
- 3 - 10 %
- ⬡ 10 - 30 %

Type of Emission Source

- coal-fired electricity generation
- waste incineration
- manufacturing
- metallurgical
- other fuel combustion

1000 0 1000 2000 Kilometers



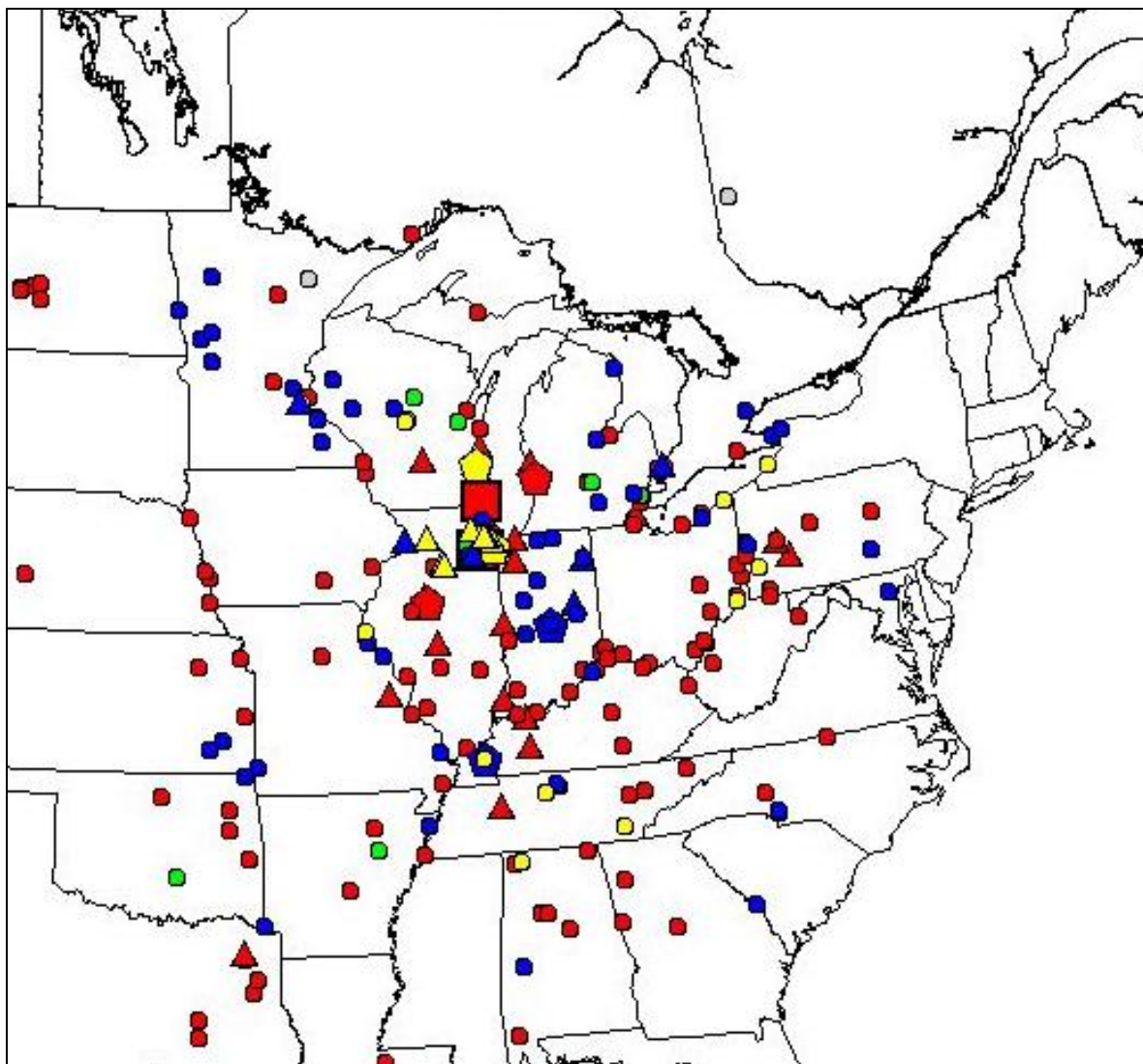
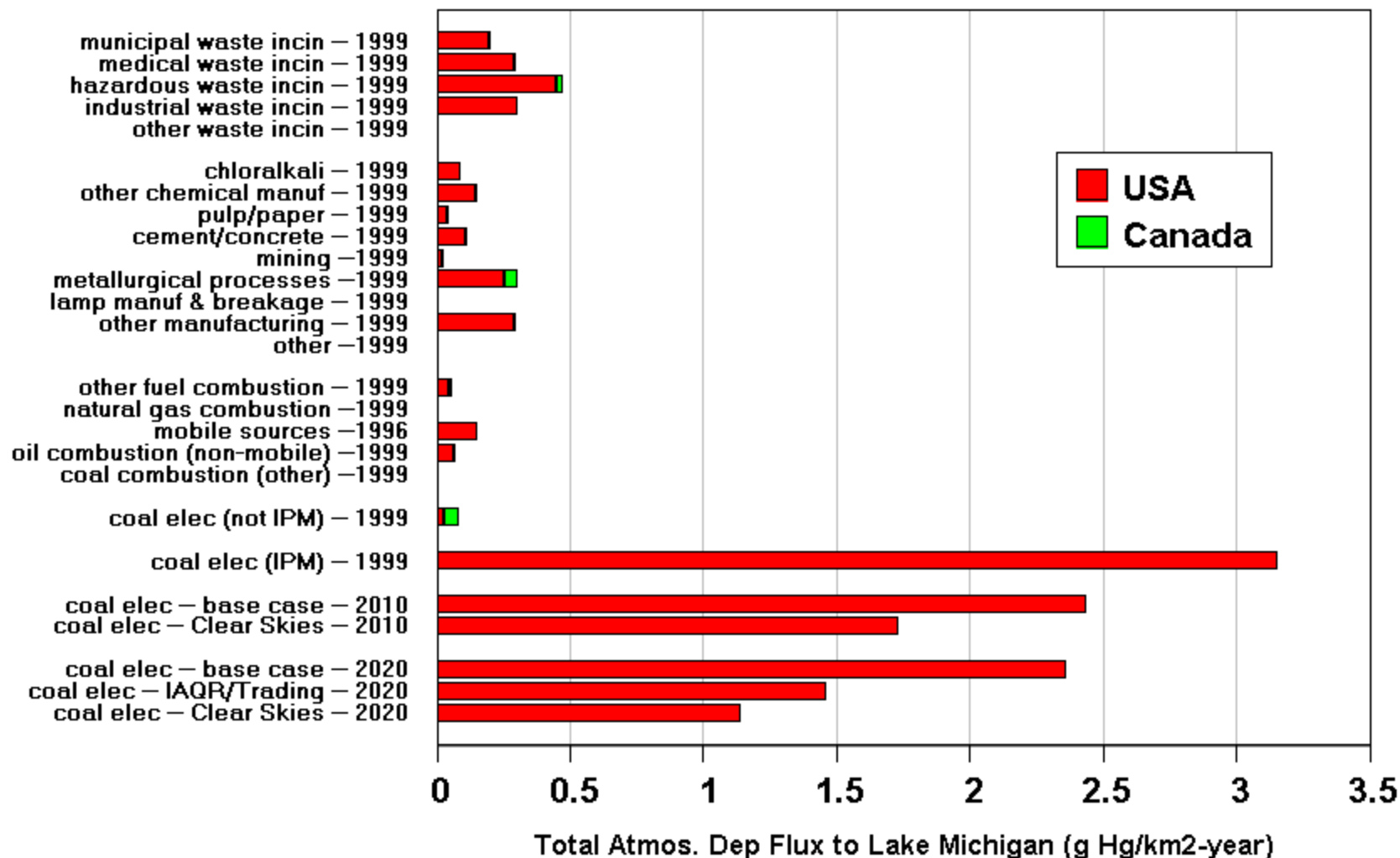
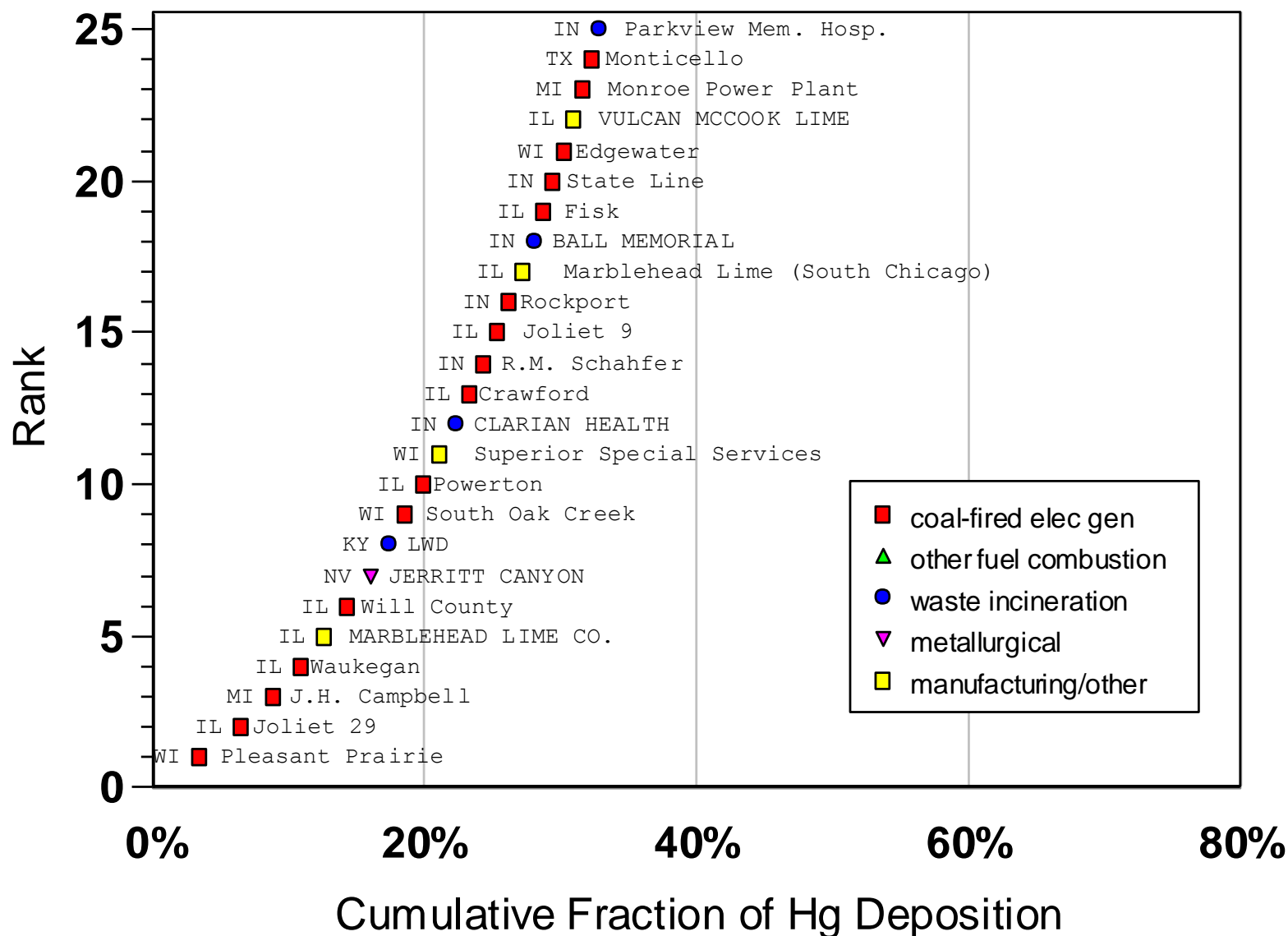


Figure 44. Largest modeled contributors to Lake Michigan (close-up).
(same legend as previous slide)

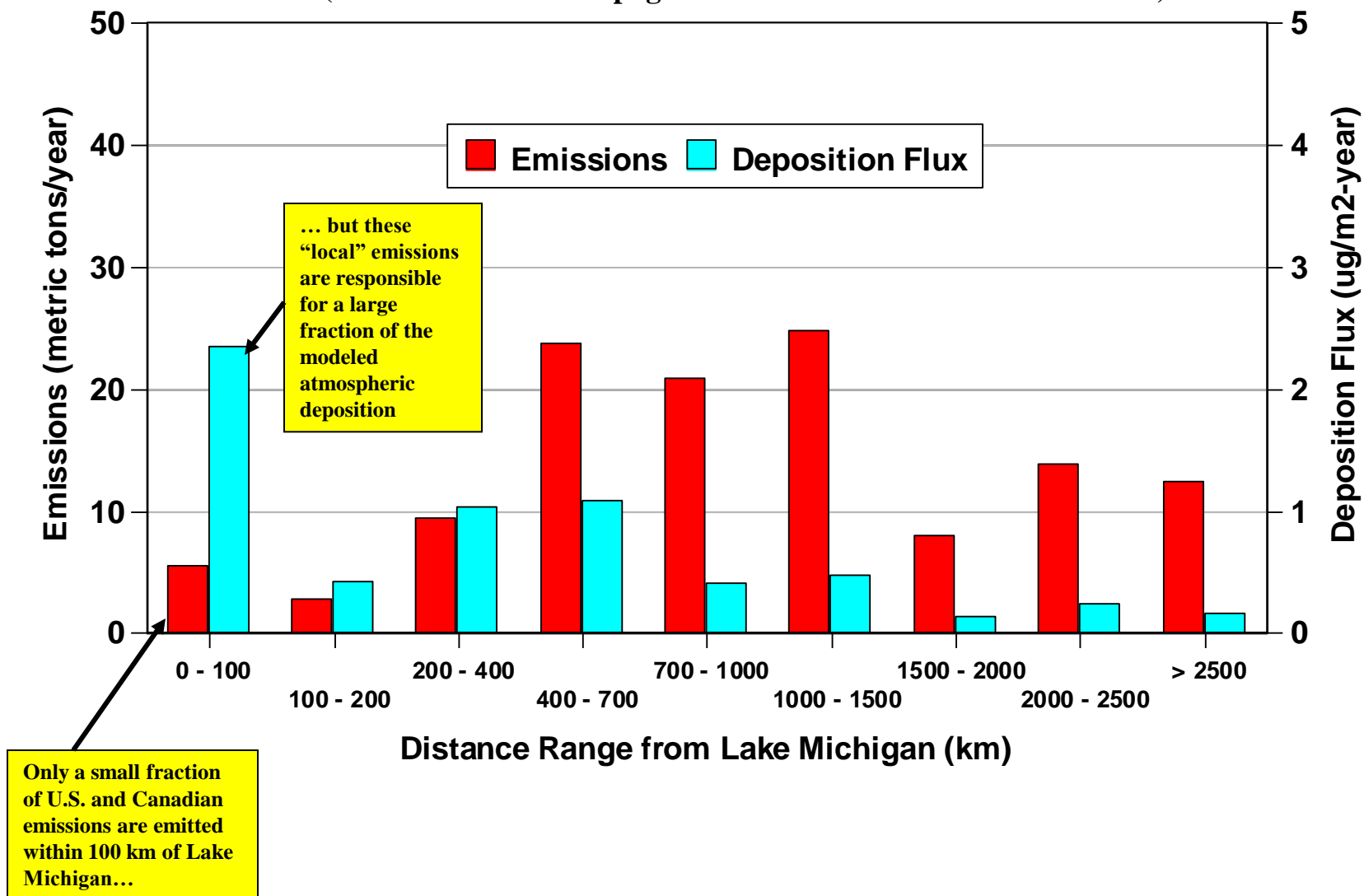
Atmospheric Deposition Flux to Lake Michigan from Anthropogenic Mercury Emissions Sources in the U.S. and Canada



Top 25 modeled sources of atmospheric mercury to Lake Michigan (based on 1999 anthropogenic emissions in the U.S. and Canada)



Emissions and deposition to Lake Michigan arising from different distance ranges (based on 1999 anthropogenic emissions in the U.S. and Canada)



A satellite image of the Indian Ocean, showing the surrounding landmasses of Africa, Asia, and Australia. The ocean is a deep blue, with white clouds visible over the land and sea. The word "Thanks!" is written in white, bold, sans-serif font in the center of the image.

Thanks!